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CHARACTERISTICS OF A THERMIONIC CONVERTER WITH A CHLORIDE VAPOR DEPOSITED TUNGSTEN EMITTER (110) AND A NICKEL COLLECTOR

by V. C. Wilson and S. P. Podkulski

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CHARACTERISTICS OF A THERMIONIC CONVERTER WITH A CHLORIDE

VAPOR DEPOSITED TUNGSTEN EMITTER (110) AND

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ABSTRACT

A chloride vapor-deposited (110) tungsten emitter with a vacuum work function of 4.96 eV was built into a 0.005-inch-spaced converter with a nickel collector. The I-V curves for emitter temperatures of 1673°K to 2153°K are presented. The output power versus emitter temperature is compared with three other similar converters. This converter yielded almost the same output power as a converter with a single-crystal (110) tungsten emitter, 0.002-inch spacing and a molybdenum collector. The vapor-deposited (110) tungsten surface was found to be extremely stable. The collector work function was observed to decrease when the device was left at room temperature for one month.

FOREWORD

The research described herein was conducted by the General Electric Research and Development Center under NASA Contract NAS 3-8511. The NASA Project Manager was Mr. J. F. Mondt, Direct Energy Conversion Division, Lewis Research Center. The report was originally assigned General Electric document No. GESP-9001.

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SUMMARY

This topical report* represents one of a series of reports to be prepared under NASA Contract NAS 3-8511, Task III, "Investigation of the Effect of Electrode Materials, Surface Treatment, and Electrode Spacing on Thermionic Converter Performance."

The converter of this report had a fixed electrode spacing of 0.005 inch. The emitter was chloride vapor-deposited tungsten with the (110) crystal planes parallel to the surface. The collector and guard ring were nickel.

Four emitters were prepared. The two that x-ray diffraction indicated were highly oriented with the (110) crystal planes parallel to the emitter surface, grew large grains when heated to 2500°C. The heat treated well-oriented surfaces had vacuum work functions of 4.95 eV. Measurements on single crystal (110) planes vary from 4.92 to 5.25 eV.

Since heat treatment at 2500° converts most of the surface to the (110) crystallites and since further heating at the converter operating temperature increases the area of (110) surface, one expectes the (110) surface to be extremely stable during converter operation.

Initially, the collector work function was 1.61 eV. After a month at room temperature, it was 1.44 eV. After this change in collector work function, the converter was operated with the emitter temperature, T_E, from 1673°K to 2153°K; collector temperature, T_C, from 827°K to 1065°K; and the cesium reservoir temperature, T_{Cs}, from 533°K to 653°K.

A calculated efficiency at 2155°K for the emitter was 23%. Because of the shape of the envelope of the load lines at various cesium pressures at

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*The data in this document were first reported at the Second International Conference on Thermionic Electrical Power Generation at Stresa, Italy, May 27-31, 1968 (see ref. 1).

this emitter temperature, the plot of the efficiency versus current density had a broad maximum with efficiencies above 22% from 10 to 40 amperes/cm^2.

The output power of this converter with the polycrystalline (110) tungsten emitter, a nickel collector and a 0.005-inch spacing was compared with the output from a converter operated by Athanis and Van Someren[1] which had a single crystal (110) tungsten emitter, a molybdenum collector, and a 0.002-inch spacing. The two converters had nearly identical output powers over a wide range of emitter temperatures and current densities.

Although the converter was only operated for about 40 hours, there was no indication of a reduction in output power with operation. Photo-micrographs of the central area taken before and after operation of the emitter showed the exact grain structure and no change in surface appearance.

The waste heat from the collector was removed by radiation and the collector temperature controlled by an auxiliary electric heater. At high emitter temperature and high current densities, the radiation cooling was not sufficient and the collector rose to 1065°K which is considered above the optimum collector temperature. This reduced the observed output power somewhat at high emitter temperatures.

INTRODUCTION

This study is part of a program to compare the characteristics of thermionic converters with various emitter and collector materials and surfaces under Task III of contract NAS 3-8511. Table 1 indicates the emitter material, collector material and spacing for the first eight converters of this series. Item 9 is the converter reported here. The measured performances are documented in the references indicated in the table.

Table 1.

	Emitter	Collector	Spacing (Inches)
(1)	Polycrystalline W[2]	Ni	0.005
(2)	Polycrystalline W[2]	Ni	0.005
(3)	Polycrystalline W[3, 4, 5]	Ni	0.002
(4)	Polycrystalline W[5, 6]	Ni	0.002
(5)	Polycrystalline W[6]	W	0.002
(6)	W-25 w/o Re[7]	Ni	0.005
(7)	Polycrystalline W[8, 9]	Nb	0.001 to 0.020
(8)	Vapor Dep. (100) W, (110) Etch[10]	Nb	0.001 to 0.020
(9)	Vapor Dep. (110) W, (110) Etch	Ni	0.005

The converter of this report had a fixed electrode spacing of 0.005 inch. The nickel collector was surrounded by a nickel guard ring. A description of the converter was given in reference 3. The choices for emitter and collector materials were made for the following reasons. The (110) crystal plane of tungsten was chosen for the emitter for two reasons: (1) it is the close-packed plane and it yields a higher electron emission at intermediate cesium vapor pressures than the other planes of tungsten;[11] and (2) the close-packed plane is least susceptible to thermal etching--at normal, emitter temperatures should be stable with time. For the collector, nickel was chosen because previous tests[6] indicated nickel covered with cesium has been a stable, low-work function surface.

In the future program, because of the anticipated stability and uniformity of the tungsten (110) surface, this surface will be used as a reference emitter surface to compare various collector surfaces. The next collector to be tested is niobium.

EMITTER SURFACE CHARACTERIZATION

The emitter used for this test was one of four--each was a 1/4-inch-thick polycrystalline tungsten disk, General Electric Company #115 weldable grade, coated with a 0.020-inch-thick layer of (110) tungsten by the chloride vapor-deposition process. The vapor-deposited surface of each emitter was ground flat, mechanically polished and electropolished.[12] Table 2 shows the result of x-ray tests on these four emitters.

Table 2.

Emitters	1		2		3		4	
Angle, Degrees	CPS	I/I_o	CPS	I/I_o	CPS	I/I_o	CPS	I/I_o
0	24,770	100	19,750	100	4,810	100	2,235	78*
1/2	19,180	77	19,570	99	3,970	82	2,950	100
1	14,550	59	16,180	82	2,770	58	2,885	98
2	7,960	32	12,210	62	950	21	1,895	64
3	6,210	25	8,970	45	620	13	1,225	42
4	5,320	21	5,500	28	500	10	545	18
5	4,010	16	3,750	19	340	7	400	14

*Fiber axis not normal to disk.

The x-ray technician made the following observations, ". . . the exposed (110) crystal planes of emitters 1 and 2 are random within the emitter plane and more highly oriented than (110) crystal planes of emitters 3 and 4. The (110) crystal planes of emitters 3 and 4 exhibit some randomness parallel to the surface of the emitter with a small degree of orientation within the plane." The emitter was set up in the normal fashion in the diffractometer to determine the orientation of the emitter surface. From all four emitters the (110) Bragg reflection was stronger than any other Bragg reflection. The incident beam covers a swath across the sample 1/4-inch wide. If there are many crystals with the (110) plane parallel to the emitter plane, the counting rate will be high. If there are only a few crystals with the (110) plane parallel to the emitter plane, the counting rate will be low. Emitters 1 and 2

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have five to ten times the counting rates of emitters 3 and 4. This is the principal indication that emitters 1 and 2 are better oriented than emitters 3 and 4.

The x-ray goniometer was set up to reflect the (110) Bragg reflection and the assembly was tipped a few degrees in a plane perpendicular to the plane containing the incident beam, the normal to the surface and the Bragg reflection. The resulting x-ray intensities as a function of the angle of this tip are given in Table 2 in CPS (counts per second). The relative intensity, I/I_0, for each emitter is given in Figure 1. These plots are not sufficient to judge which emitters are best oriented. The low counting rate for emitter 3 indicates that there are few crystallites oriented with the (110) parallel to the surface; however, the rapid fall-off of relative intensity with angle indicates that those crystallites with the (110) orientation are very closely oriented parallel to the bulk surface.

Each emitter was mounted in a vacuum bell jar (pressure $\sim 2 \times 10^{-6}$ torr with emitter hot) with the oriented surface facing a collector and guard ring. The work function of this surface was determined by electron emission from the emitter to the collector in an accelerating field for electrons. Figure 2 is a plot of work function at different temperatures for emitter 3. Initially contamination, probably oxygen, caused the work function to be high. After first heating to 2400°K and finally 1/2-hour at 2670°K, the work function settled down to 4.73 eV measured at 2200°K. Figure 3 shows the results of work function measurements for emitter 1 at different temperatures. The lower curve was taken after a heat treatment of 1 hour at 1973°K and 1 hour at 2100°K. The small change in work function as a function of emitter temperature suggests that this heat treatment cleaned up the surface. After further heating for 1/2-hour at 2670°K, the upper curve was obtained. During this high temperature heat treatment, larger crystals were grown in the tungsten. This behavior is shown in Figure 4a. The three fine lines that meet near the center of the photograph are new crystal grain boundaries. The lines which outline smaller areas are the grain boundaries of the original
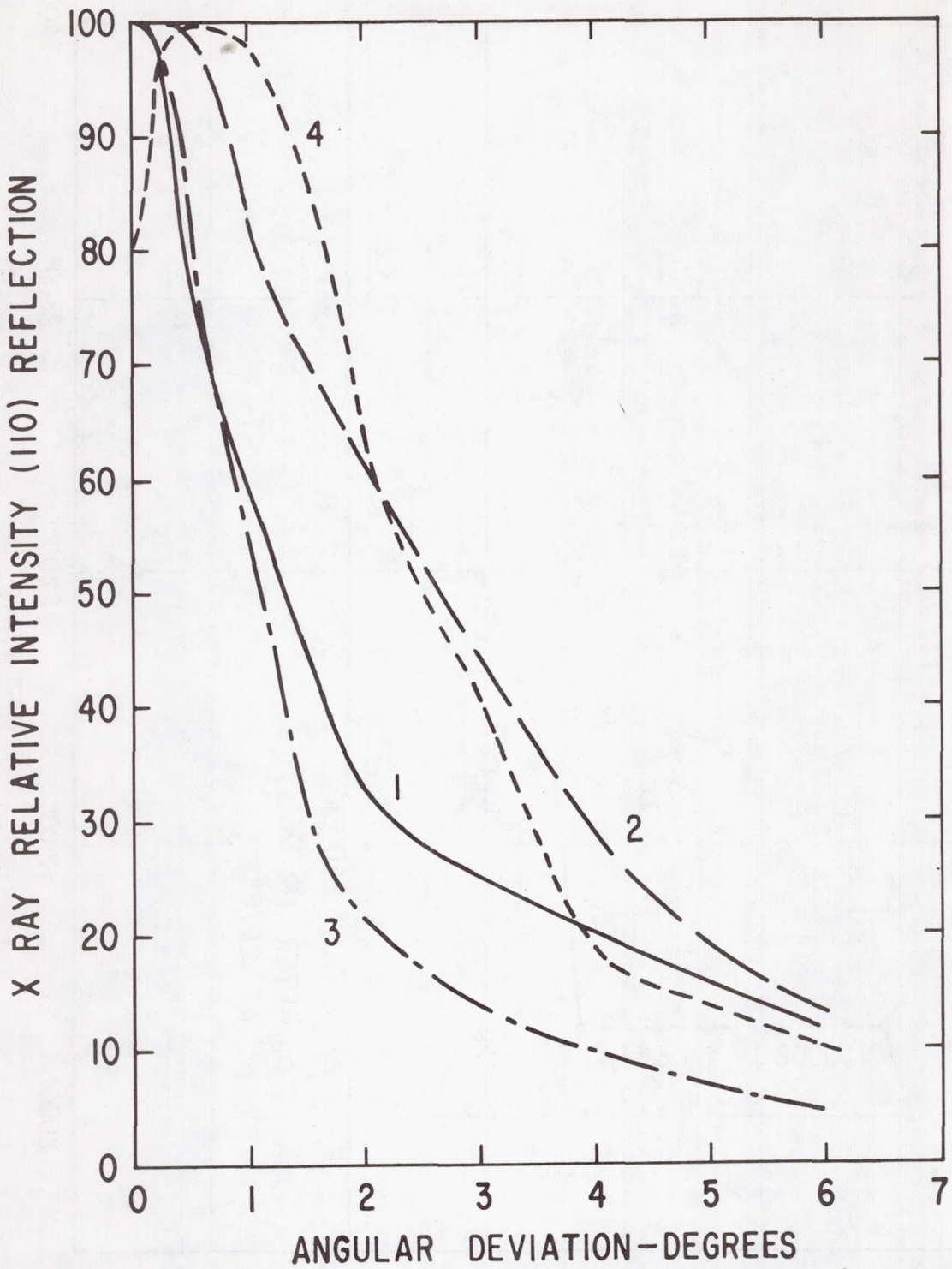
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FIGURE 1. RELATIVE REFLECTION INTENSITY FROM THE (110) CRYSTAL PLANES AS A FUNCTION OF DEGREES FROM THE NORMAL TO THE PLANE
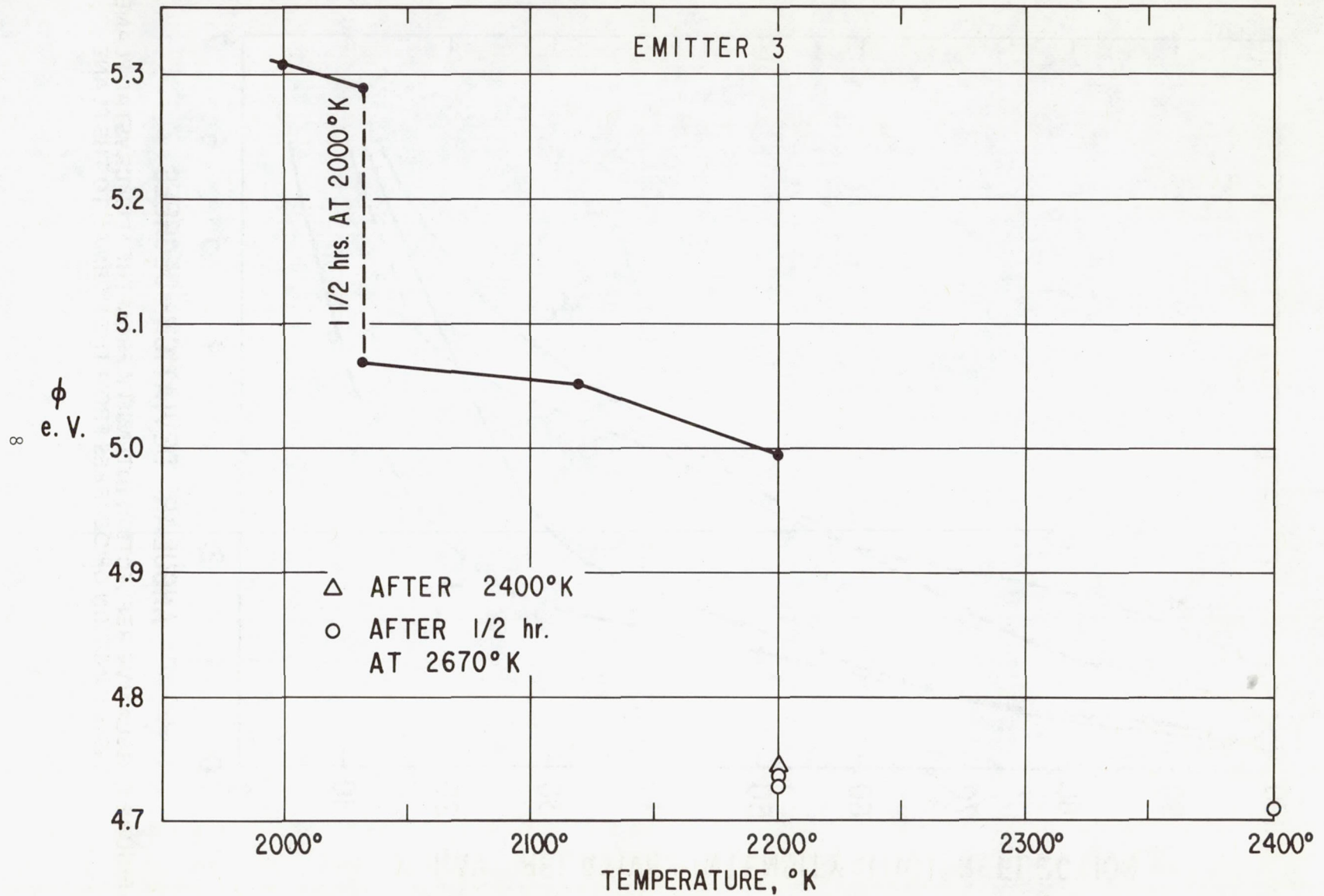
FIGURE 2. CHANGES IN WORK FUNCTION OF EMITTER 3 CAUSED BY VACUUM
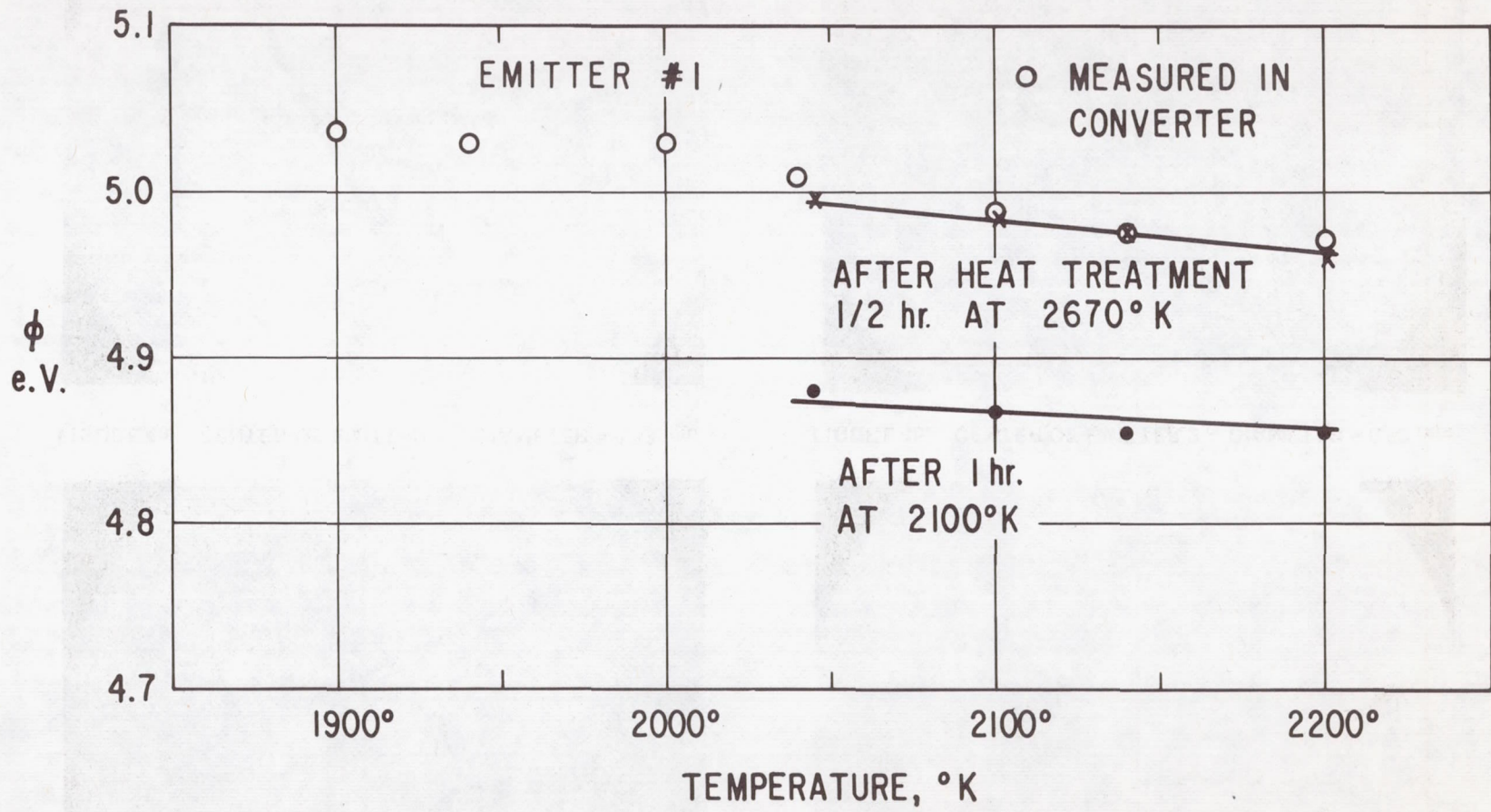HEAT TREATMENTS AT DIFFERENT TEMPERATURES

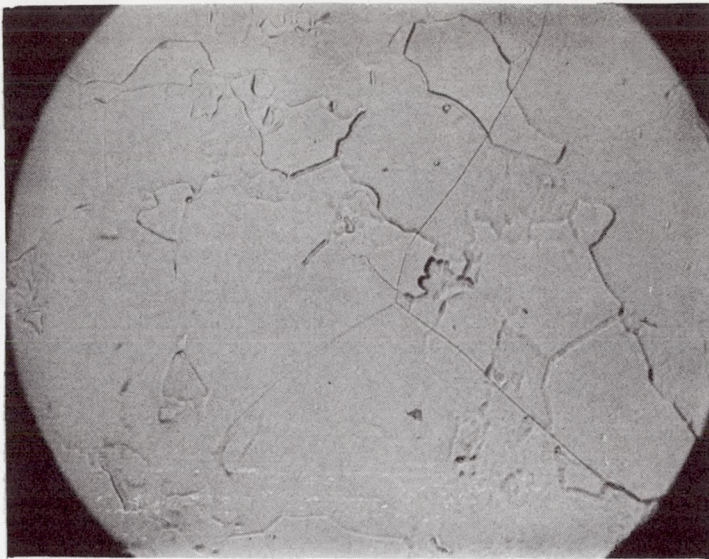FIGURE 3. INCREASE IN ϕ OF EMITTER 1 CAUSED BY (110) CRYSTAL GROWTH

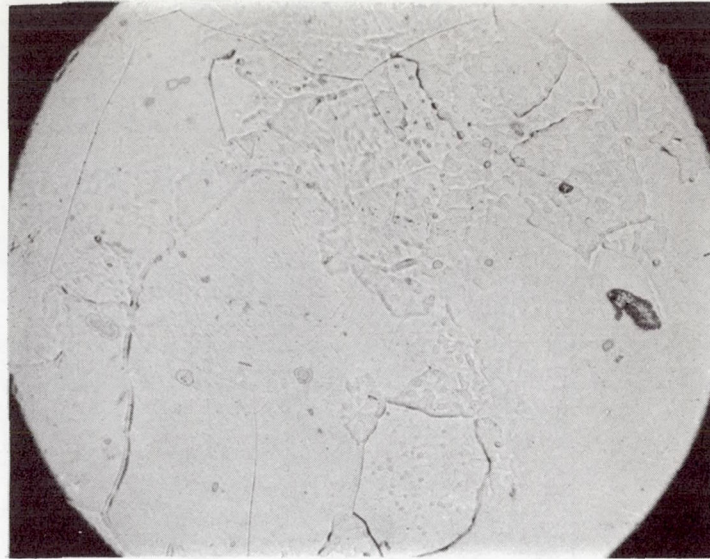FIGURE 4a. CENTER OF EMITTER 1 — DIAMETER = 0.53 mm

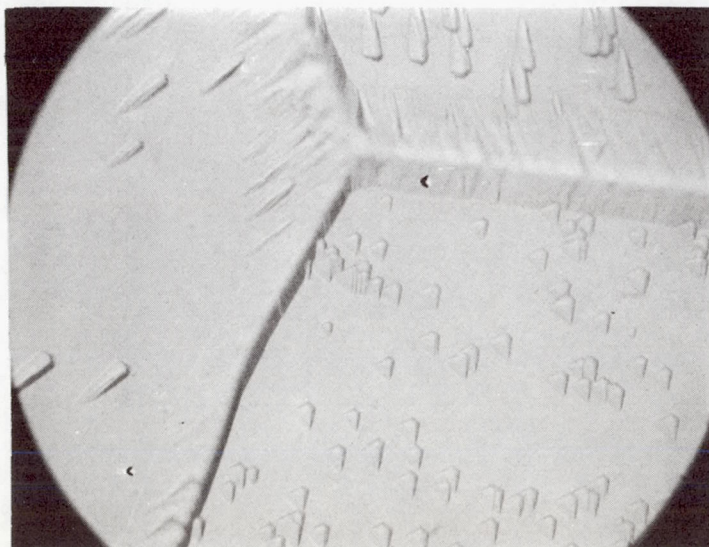FIGURE 4b. CENTER OF EMITTER 2 — DIAMETER = 0.53 mm
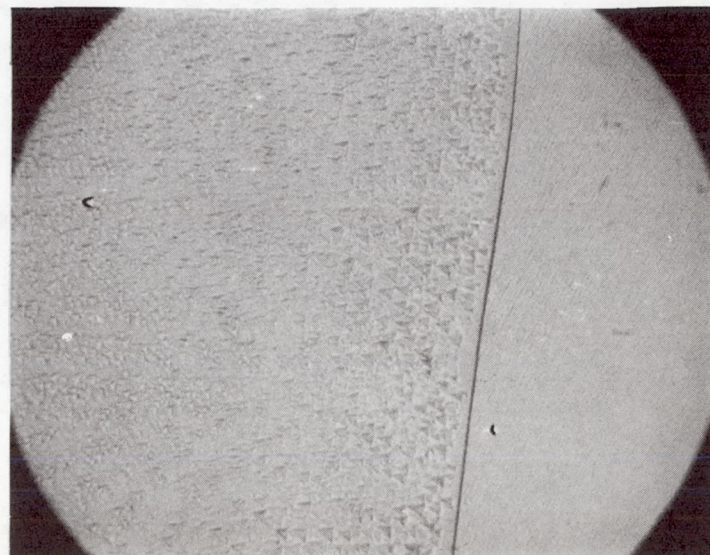

FIGURE 4c. EMITTER 3 — DIAMETER = 0.19 mm

FIGURE 4d. EMITTER 4 — DIAMETER = 0.53 mm

crystallites. These smaller areas are at slightly different elevations, like small plateaus extending above a flat plane. The difference in elevation is a micron or less. Figure 4b shows a similar region for emitter 2, Figure 4c is from emitter 3, and Figure 4d is from emitter 4. The etch pits in Figures 4c and 4d show that emitters 3 and 4 are not well oriented with the (110) planes parallel to the surface.

An electroetch of 1 minute at 2 volts produced a surface with appearance similar to a heat treatment of 1/4-hour at 2500°C except that during a heat treatment the 110 grains grew at the expense of the misoriented grains. This effect is shown in Figures 4e and 4f by a difference in magnification. In general, further heating of emitter 1 caused the surface to approach a continuous (110) surface (shown in Figure 4g) and the work function increased from 4.87 eV to 4.96 eV.

Since heat treatment at 2500°C converts most of the surface to the (110) crystallites and since further heating at converter operating temperatures increases the area of (110) surface, one expects the (110) surfaces to be extremely stable during converter operation.

The work function of the emitters at 2200°K is given in Table 3. Emitter 1 was selected for this converter.

Table 3.

Emitter	ϕ (eV) at 2200°K
1	4.96
2	4.95
3	4.73
4	4.67

L. Yang and R. G. Hudson[13] have also observed a correlation between the degree of orientation of the chloride deposited tungsten and the magnitude of the work function. They prepared a duplex structure with (110)-oriented fluoride tungsten base, and report ". . . stable vacuum work
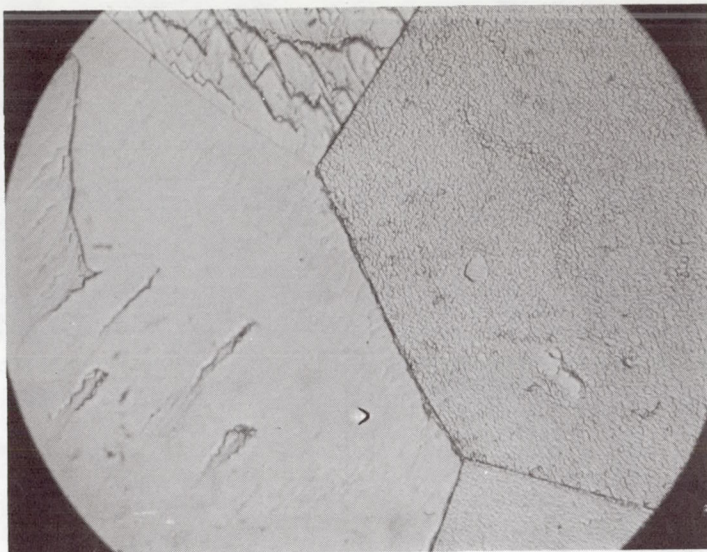
FIGURE 4e. EMITTER 2 AFTER ELECTROETCH; 2 VOLTS
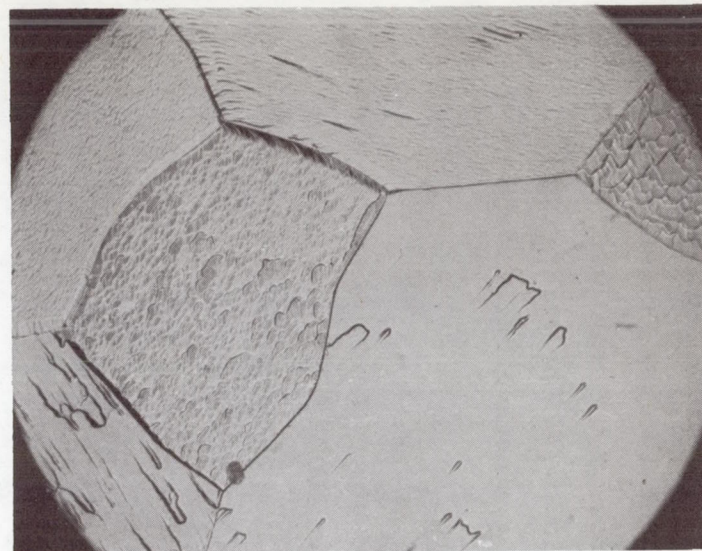FOR 1 MINUTE — DIAMETER = 0.19 mm

FIGURE 4f. EMITTER 2 AFTER ¼ HOUR AT 2500°C
DIAMETER = 0.53 mm

FIGURE 4g. TYPICAL AREA OF EMITTER 1 AFTER HEAT TREATMENT —
DIAMETER = 0.19 mm

function at 2073°K". They report, "Thermal etching in vacuum up to 2673°K
does not change the vacuum work function of either the chloride or the fluoride
tungsten deposit."

CONVERTER PERFORMANCE TESTS

After outgassing the converter, the work function of the emitter was again measured. The circles on Figure 3 show the results of these measurements. After admitting cesium, the collector work function was measured. initially the nickel collector was heated to 806°K and the cesium reservoir was heated from 491°K to 523°K. Under these conditions, it was impossible to measure the work function of the collector because the converter went into a discharge before the collector emission current reached saturation. The collector was then cooled to 780°K and the cesium reservoir temperature varied from 414°K to 518°K. In this range the minimum collector work function was 1.61 eV at T_{Cs} = 424°K, corresponding to a T/T_{Cs} of 1.84. The converter was then operated at about 1700°K emitter temperature and the performance indicated that it was not a particularly high-output converter.

The converter was not operated again for a month. Figures 5 through 10 show the output characteristics of the converter at different emitter temperatures. The envelopes of these families of curves are shown in Figure 11. The method of taking the data is described in an earlier report.[9] One measurement refinement was made to speed up the collection of data. Formerly, in order to measure the average a.c. (used in calculating the heat flux through the emitter) the load resistor was adjusted so the emitter temperature would stay constant when the a.c. drive was turned on or off. This would prove that the d.c. and average a.c. were the same. The d.c. was then observed. In testing this converter, a vacuum tube voltmeter was used to measure the average alternating current (not the root mean square) and this current was used to estimate the average electron cooling.

DISCUSSION OF RESULTS

Figure 12 compares the output voltage and the power of this converter at 20 amp/cm^2 output current with two other converters. All three had nickel collectors and 5-mil spacing. The two lower curves represent converters with polycrystalline tungsten and polycrystalline rhenium emitters.
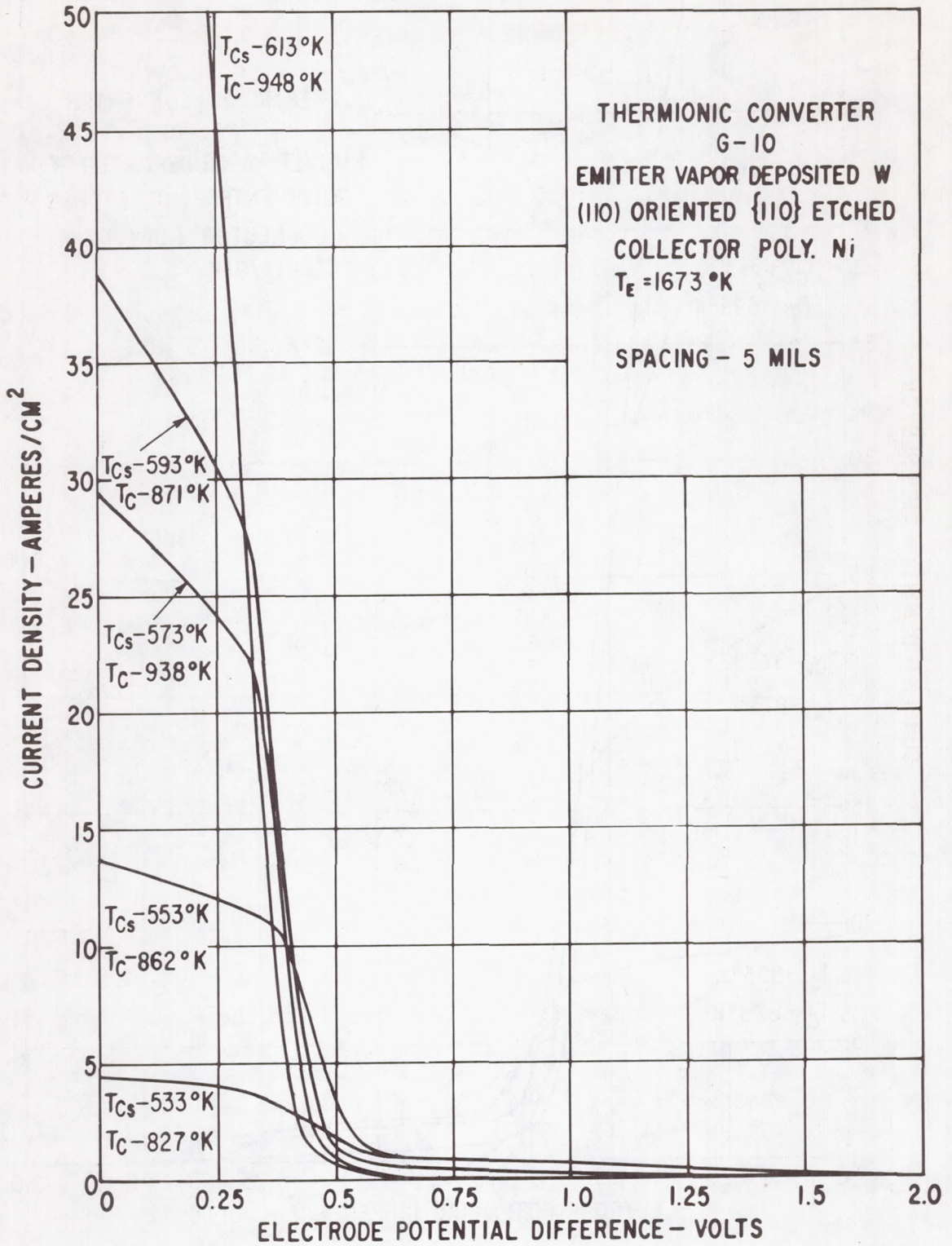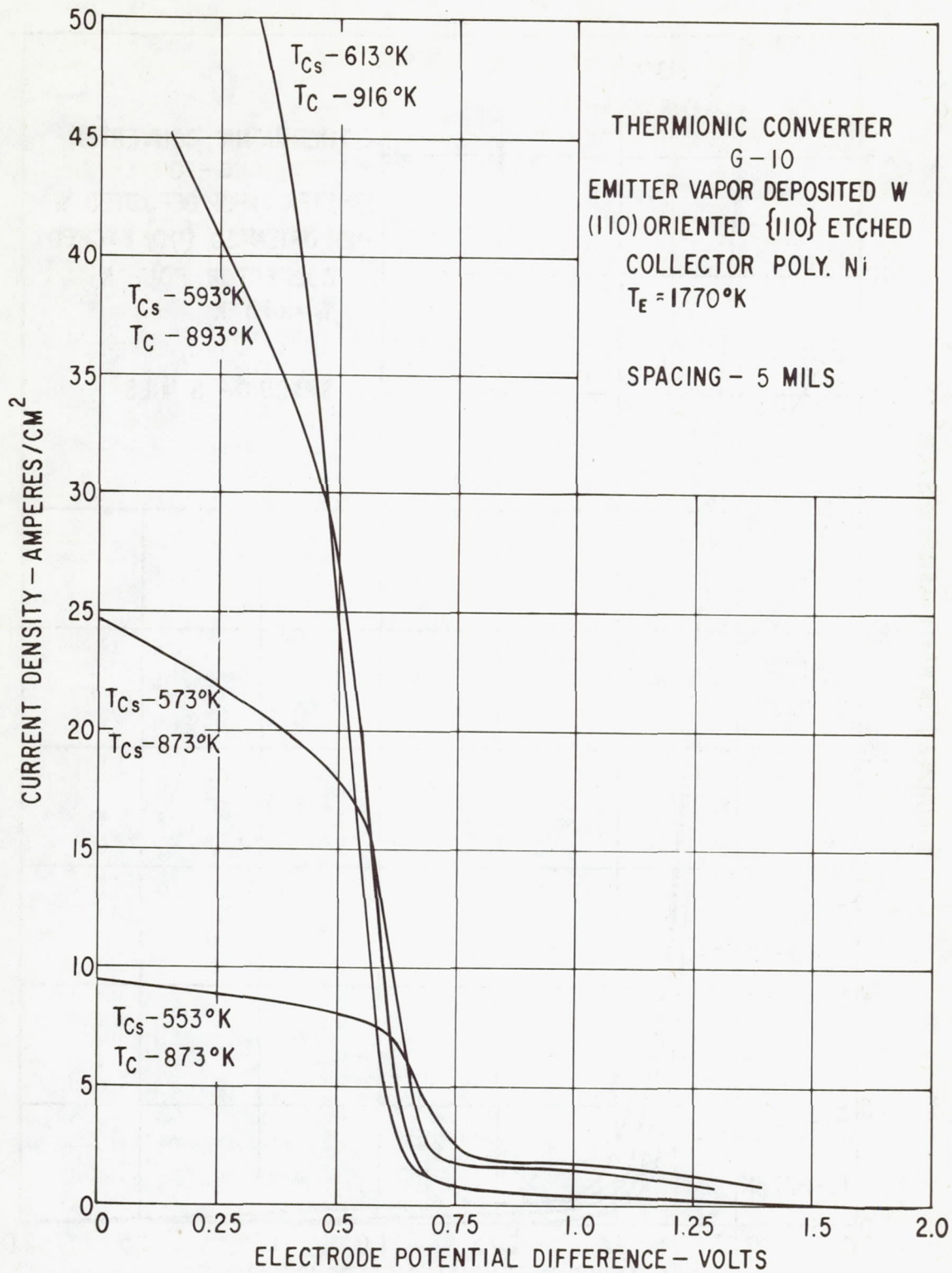
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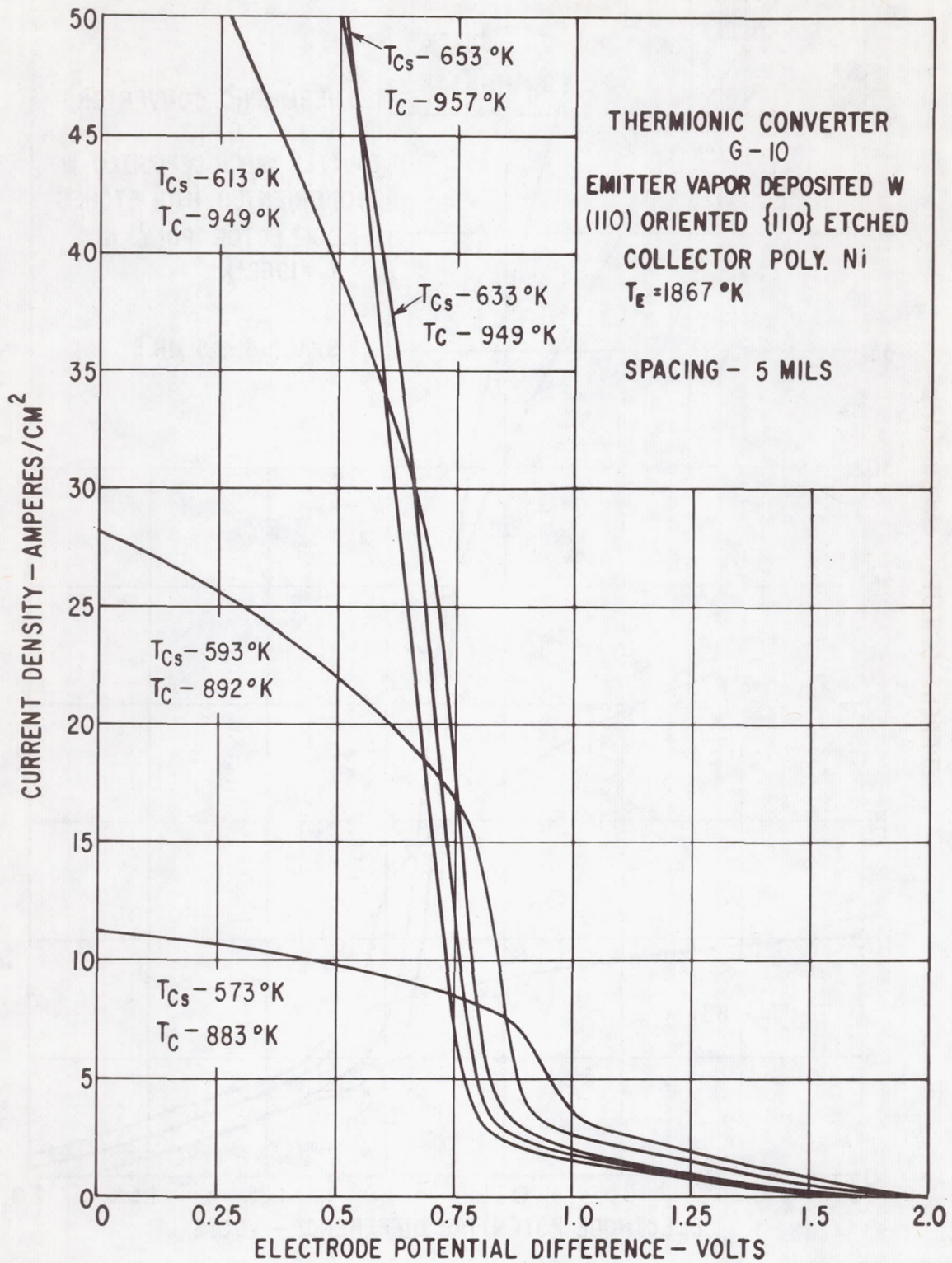FIGURE 5. LOAD LINES AT T_E – 1673°K

FIGURE 6. LOAD LINES AT T_E — 1770°K

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FIGURE 7. LOAD LINES AT T_E – 1867°K

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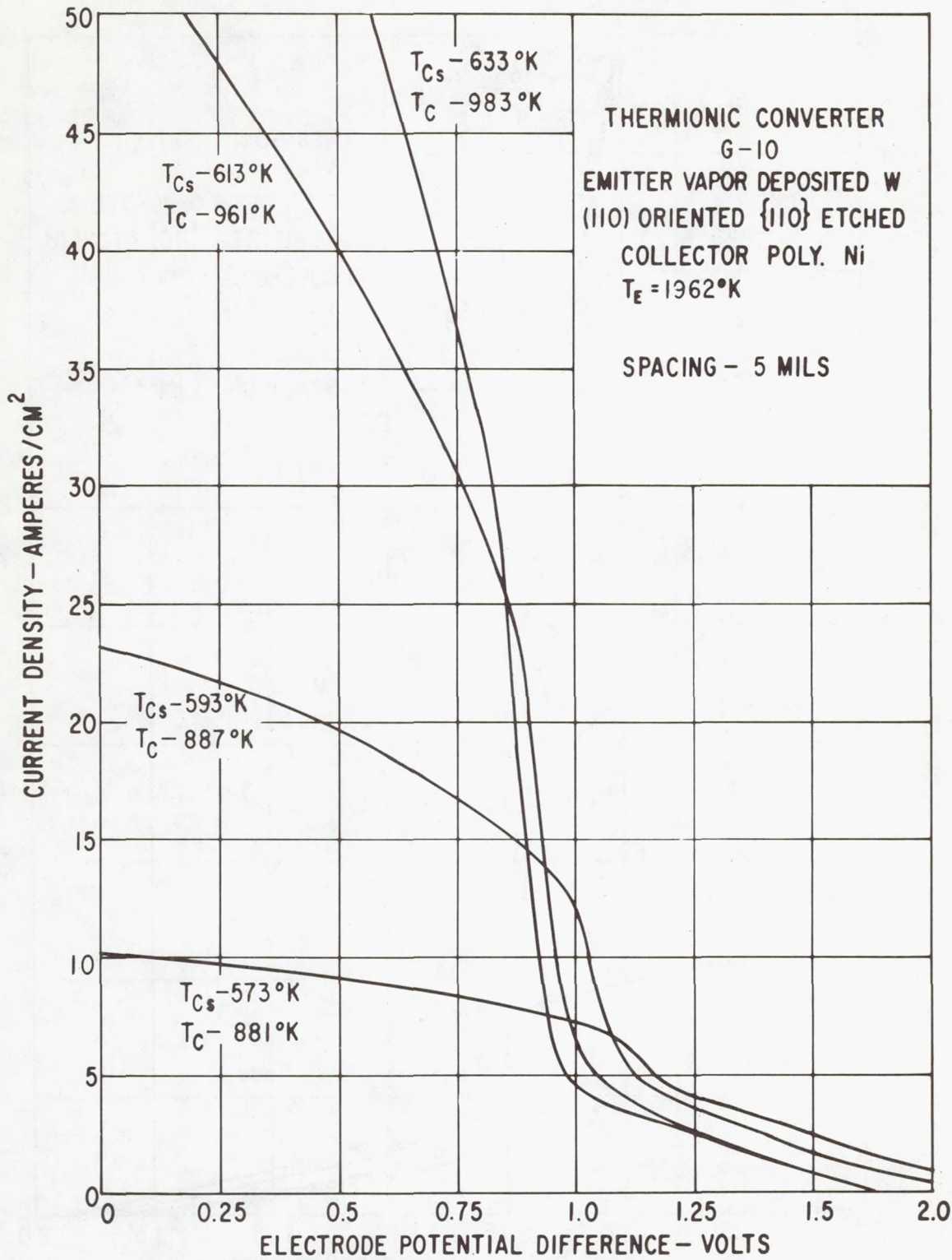
FIGURE 8. LOAD LINES AT T_E – 1962°K
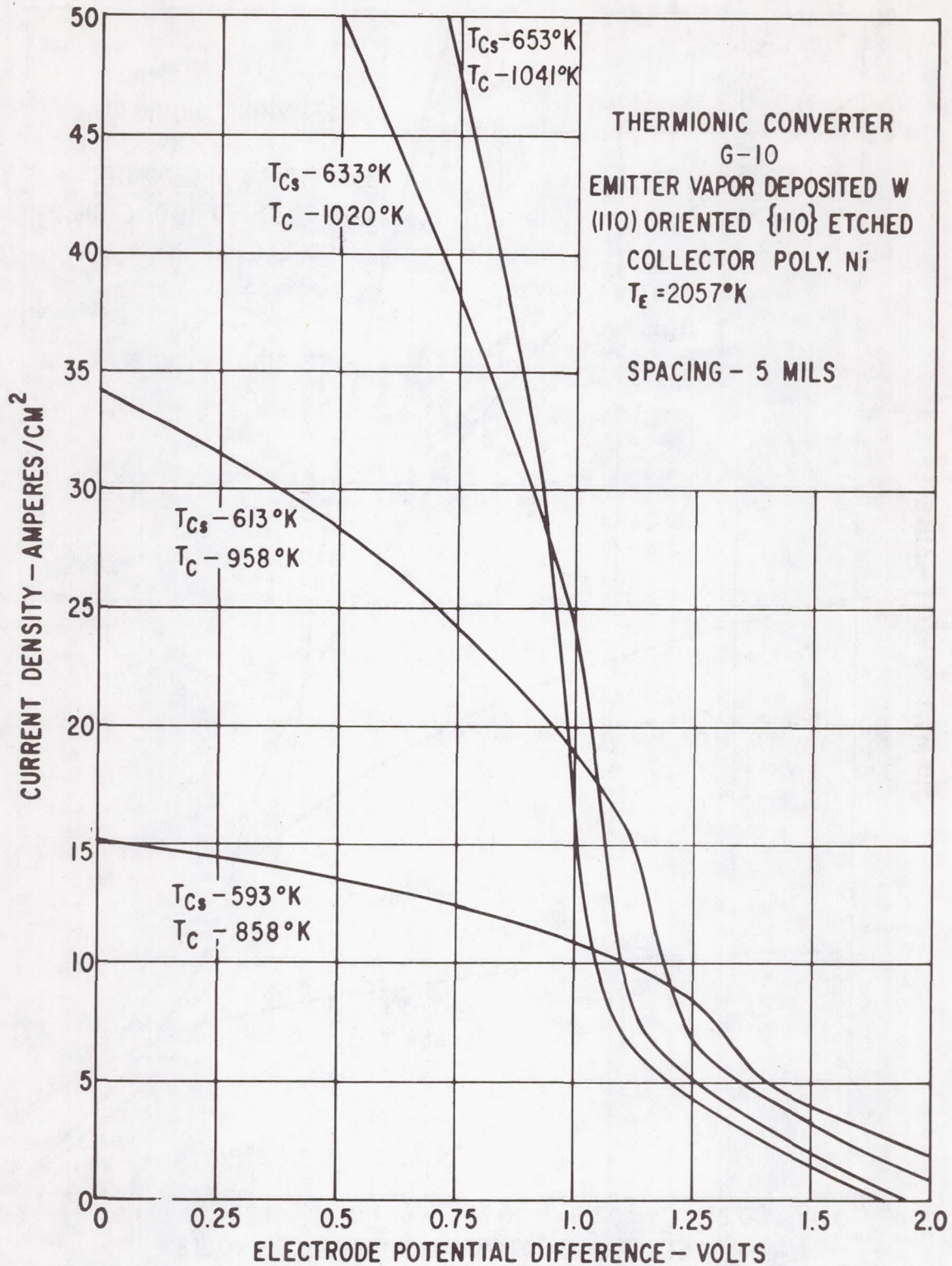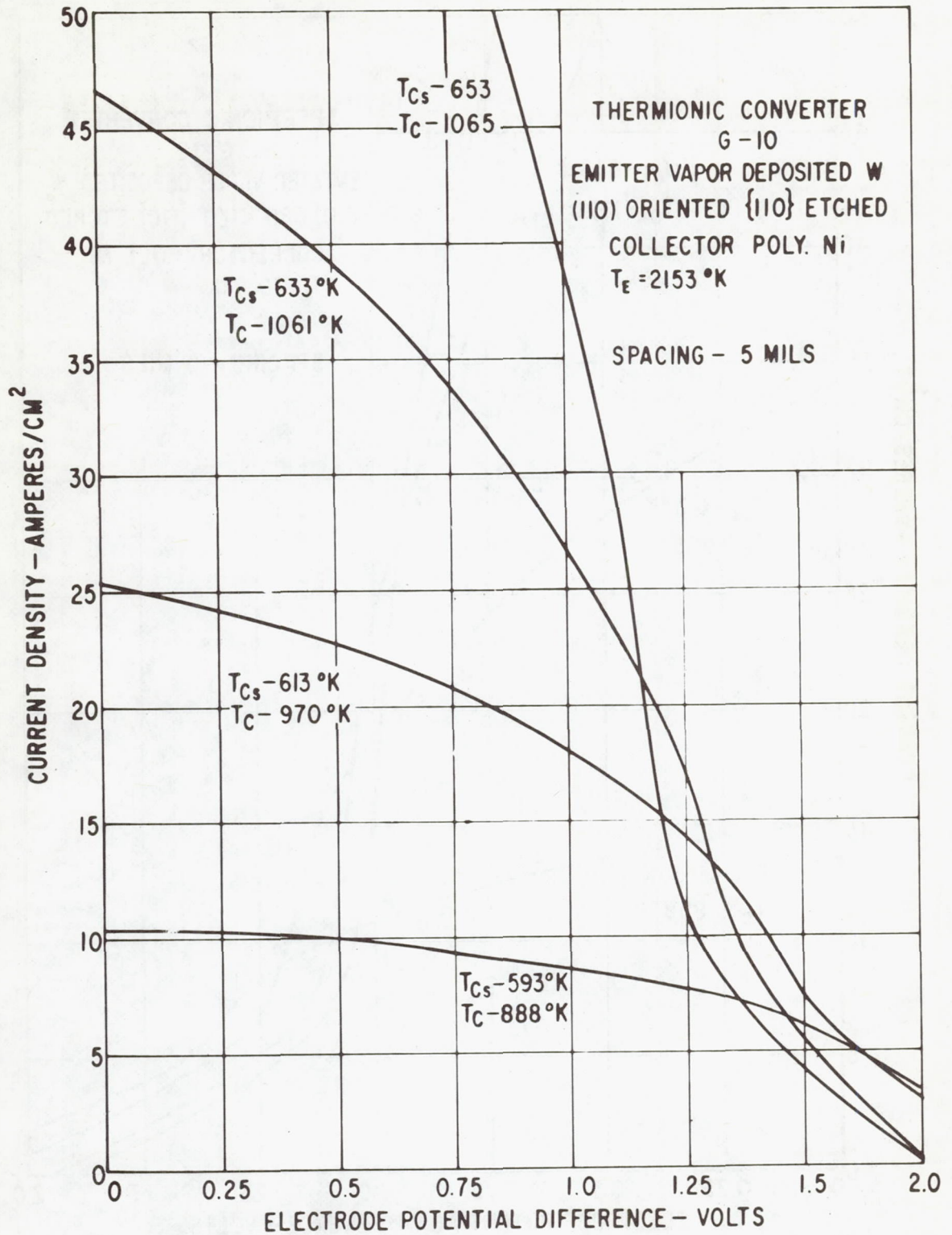
FIGURE 9. LOAD LINES AT T_E — 2057°K
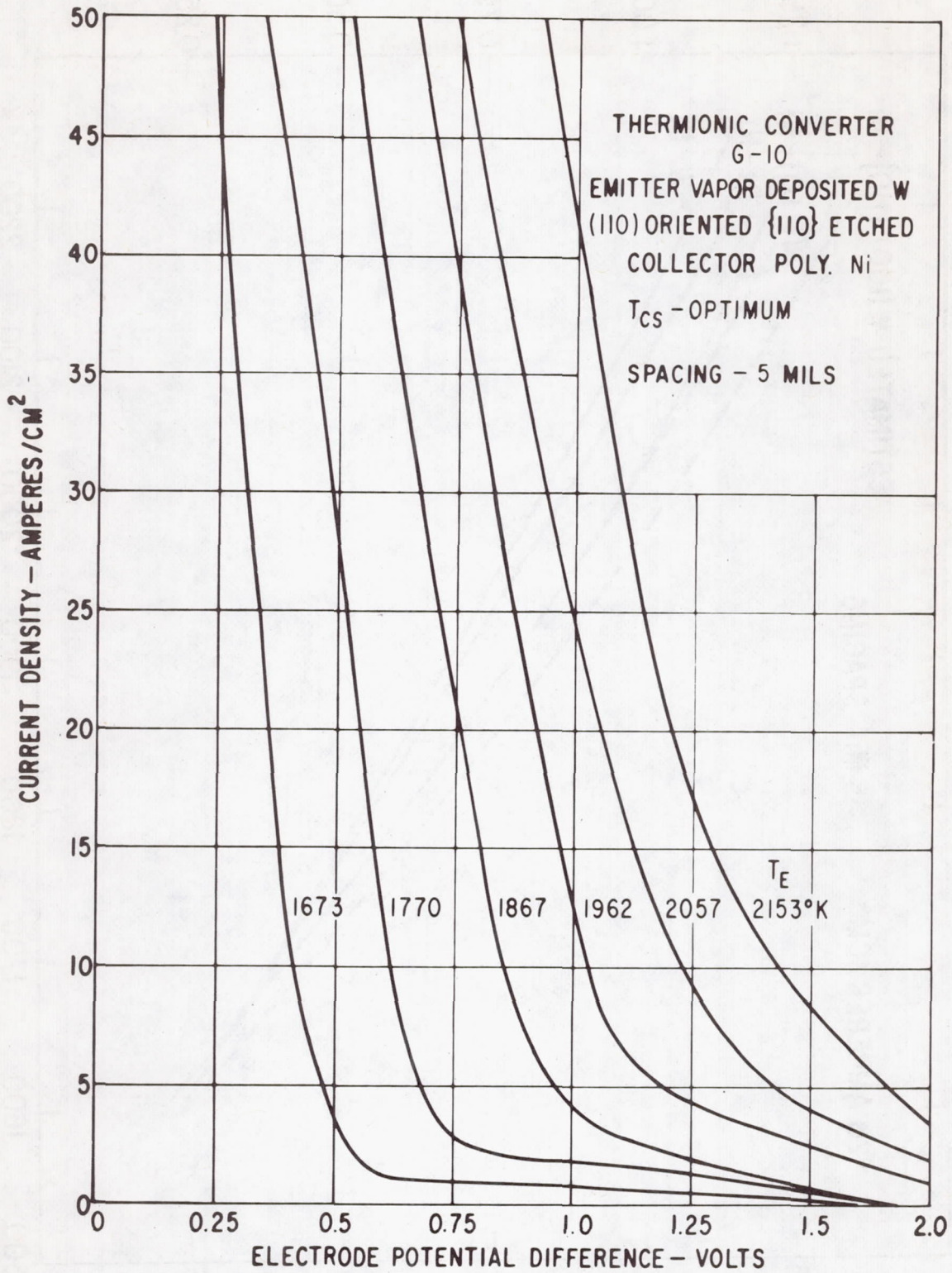
FIGURE 10. LOAD LINES AT T_E – 2153°K

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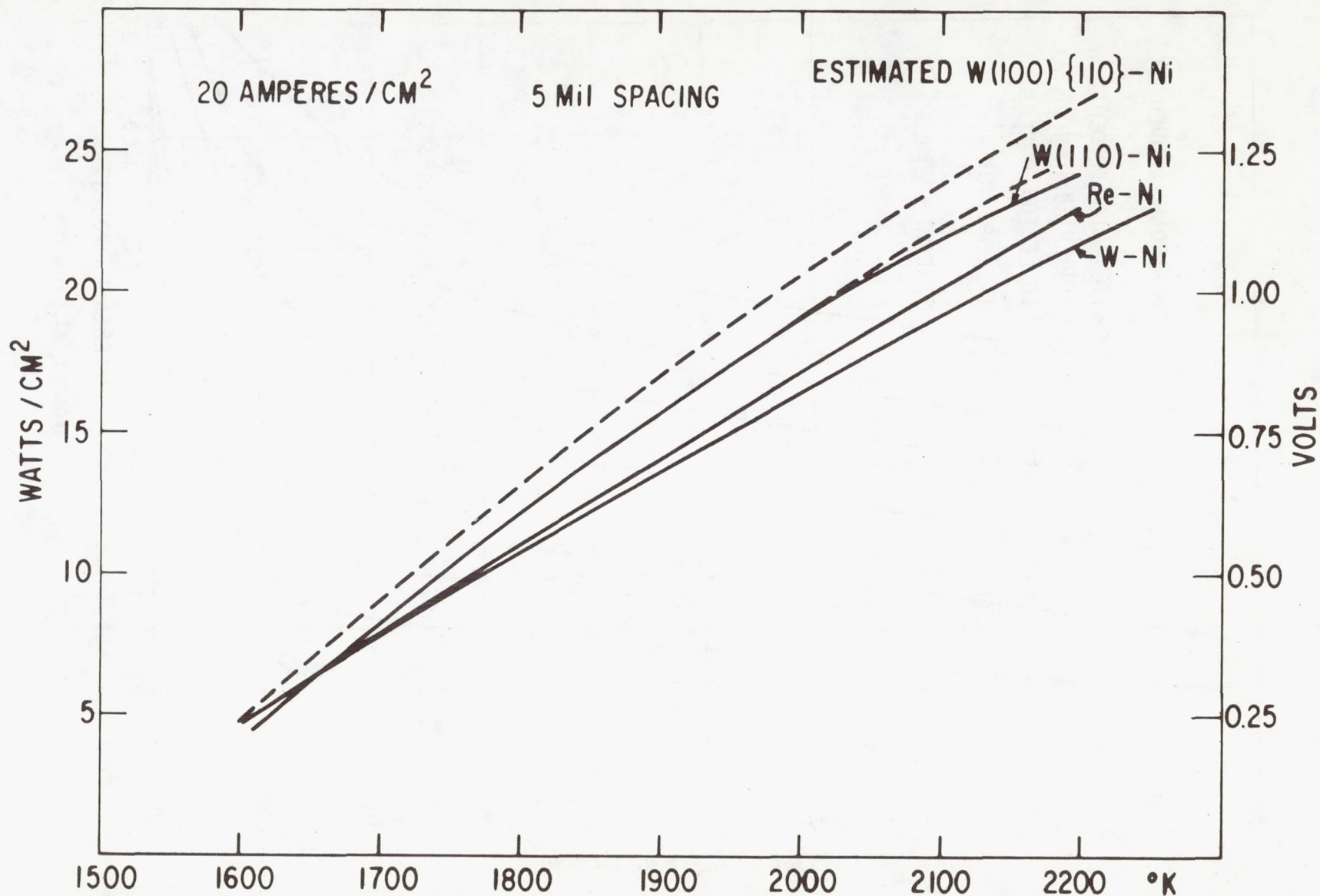FIGURE 11. OUTPUT VERSUS T_E, T_{Cs} OPTIMIZED

FIGURE 12.   COMPARISON OF OUTPUT AT 20 AMPS/CM^2 FOR THREE CONVERTERS
WITH NICKEL COLLECTORS AND 5-MIL SPACING

The upper solid curve represents the output for this converter. In this converter design, the heat from the collector is removed by radiation. This arrangement somewhat limits collector temperature control. As may be seen from Figure 10, at T_E = 2153°K, the collector ran at 1061°K for the curve that gave the optimum output at 20 amps/cm². We believe that maximum output at emitter temperature of 2153°K would be at a lower collector temperature. The dashed extension of the curve in Figure 12 is an estimated output of this converter if the collector could be kept cooler.

The preceding report[12] described a converter with a niobium collector and a tungsten emitter oriented with the (100) planes parallel to the surface but etched to expose the (110) planes. We have estimated that a converter with a nickel collector has about 0.1 volt more output potential at 20 amps/cm² current than a converter with a niobium collector. The upper dashed curve in Figure 12 includes this 0.1 volt and represents the anticipated output from a W(100){110} - Ni converter.

It is apparent from Figures 5 through 12 that the converter reported here is a high performance converter. In fact, if the collector work function is not less than the 1.61 eV initially measured, it is difficult to explain the excellent output voltages. With the emitter at 2070°K, the bias on the a.c. circuit was reduced so that the sweep would go in the reverse direction. With the collector at 883°K, 907°K, and 931°K, the load lines in the quadrant, where the converter is drawing electron current from the collector to the emitter, showed slight plateaus before going into a discharge mode. Assuming these plateaus represented the saturation electron emission from the collector, the calculated collector work function was 1.46 eV, 1.46 eV, and 1.475 eV, respectively.

During this measurement, a phase shift in the 60-cycle circuit was noted which caused a somewhat distorted load line on the x-y recorder. While trying to reduce the phase shift between the impressed voltage and observed current, it was noticed that the thermionic converter was not

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responding properly to changes in the cesium reservoir temperature. The cesium was being lost from the converter due to an envelope leak. The emitter was cooled and several measurements were made of the saturation emission from the collector at various temperatures before the cesium was lost. Eight measurements gave work function values ranging from 1.42 to 1.45 eV, with an average of 1.435 eV. Since the converter was losing its cesium and probably did not have liquid cesium in the cesium reservoir, there is no correlation between the cesium bath temperature and cesium pressure. Therefore, T_C/T_{Cs} ratios cannot be determined for these work function values. However, these measurements definitely show that the work function of the nickel collector decreased while the tube was left at room temperature for a month.

In another experiment[6] a similar result was observed with a tungsten coating on a nickel collector. Immediately after evaporating tungsten onto the nickel, the minimum work function was measured to be 1.72 eV. After the tube remained for three weeks at room temperature, the minimum work function was found to be 1.47 eV. In both cases during the initial few hours operation, there was no apparent change in converter performance. The changes appeared to occur while the converters were left for a few weeks at room temperature. One possible hypothesis is that the low work function surface is a semiconducting compound composed of the atoms of nickel or tungsten, oxygen and cesium. In the converters that have been carefully cleaned and contain cesium, the oxygen pressure is so low that it takes a few days for a monolayer of oxygen to accumulate on the collector surface. The Ni-O-Cs or W-O-Cs compound is formed at room temperature or possibly the next time the collector is warmed. This explanation is very tentative and further observations are needed to verify this hypothesis.

One might instead postulate the high temperature operation (1061°K) of the collector cleaned up the nickel and lowered the work function of the cesium-covered collector. If this is the explanation for the improvement of the converter output, the converter would have shown progressive improvement in output with operation. The first performance tests were at $T_E = 1770^\circ$K,

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$T_{Cs} = 553^{o}K$ and $T_c = 873^{o}K$ (Figure 6). This gave an output of 0.67 volts at 5 amps/cm^2. The converter,[12] $W(100)\{110\}$- Nb, gave an output of 0.48 volts at 5 amps/cm^2 at $T_E = 1745^{o}K$, $T_{Cs} = 553^{o}K$, $T_c = 923^{o}K$, 5-mil spacing (see Figure A-9 of reference 12). Figure 11 at 1770°K also shows good output at 20 amps/cm^2. It appears as though this (110) W-Ni converter had high output immediately after remaining at room temperature for one month, and even heating the collector to 1065°K (Figure 10) did not change the low work function of the cesium coated collector.

A converter previously tested[12] had a (100) oriented tungsten emitter etched to expose the (110) planes. This treatment produced a rough surface with an area estimated to be 1.41 times the geometrical area. Since both emitters had the (110) planes of tungsten exposed, one would expect that they would have the same ability to adsorb cesium. If one tries to match load lines for the two converters with the same spacing and T_E values, it is found in general that the converter with the $W(110)\{110\}$ emitter requires a cesium reservoir temperature about 20°K higher. If one divides the current density of the previous converter by 1.41 to correct for the surface area difference, the $W(110)\{110\}$ converter still requires a cesium reservoir temperature from 7°K to 10°K higher to make the load lines match. This suggests that the $W(100)\{110\}$ emitter adsorbed the cesium slightly better than the $W(110)\{110\}$ emitter. One explanation for this is that the $W(100)\{110\}$ emitter might have had a trace of oxygen on the surface which increased the bare work function slightly and increased the cesium adsorption. Probably it would only take a small fraction of a monolayer of oxygen to make this small difference. This explanation seems plausible, because in the final processing the $W(110)\{110\}$ was heated to a higher temperature and outgassed more thoroughly than the $W(100)\{110\}$ emitter.

Figure 13 shows an efficiency estimate made at $T_E = 2155^{o}K$ using calculated input power and observed output power corrected for voltage drop in the emitter lead. The envelope of the family of load lines had a comparatively high output voltage at 10 amps/cm^2. This result gives the unusually
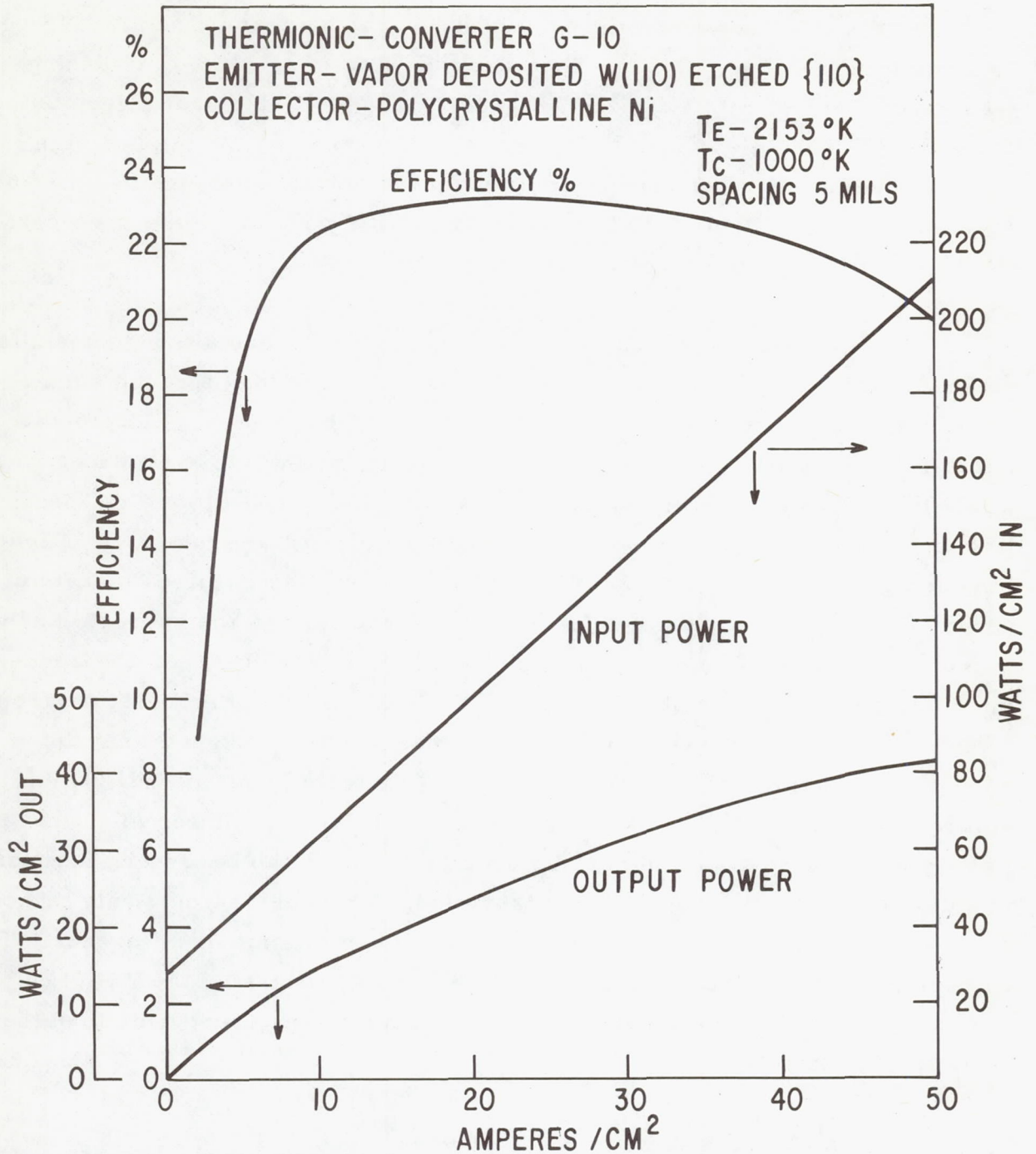
FIGURE 13.   EFFICIENCY VERSUS CURRENT DENSITY AT T_E 2153°K FOR W(110) 110 - Ni CONVERTER

high efficiency of 22% at this low current density. Notice that the efficiency curve shows a broad maximum, the efficiency varying by a factor of only 1.05 in the range 10 amps/cm^2 to 40 amps/cm^2.

POST-TEST ELECTRODE EXAMINATION

The converter was disassembled and the electrodes examined. Figure 14a shows the collector and guard. The nickel collector shows extensive recrystallization. As mentioned previously, at T_E = 2057°K the collector could not be cooled sufficiently and it was operated hotter than the guard. Also, there are 29 spots on the two electrodes that appear as though arc discharges had occurred. Probably these occurred while measuring the work function of the collector. It was mentioned previously that during collector work function measurements (T_C = 806°K) the converter went into discharges. Twelve x-y recordings show such discharges and probably additional discharges occurred when recordings were not being made. Figure 14b shows an enlargement of two of these spots.

The emitter looked almost exactly as it did when put into the converter. Figures 15a and 15b show the same area of the emitter center before and after operation. The ghosts of the original grain structure are easily identified. The new grain boundaries have moved slightly. Particularly, the right-hand end of the right-hand boundary in Figure 15b has moved down a few microns. These two pictures illustrate that this type of tungsten emitter surface is extremely stable.

PERFORMANCE COMPARISON WITH OTHER ELECTRODE MATERIALS

Test results on a thermionic converter with a single-crystal (110) surface tungsten emitter and a molybdenum collector obtained by Athanis and Van Someren have been summarized previously.[1] From discussion with Athanis, it was learned that the voltages reported are usable output voltages. To determine the electrode potential difference, the ohmic potential loss in the emitter support lead must be added to the output voltage. Also, Athanis and Van Someren did
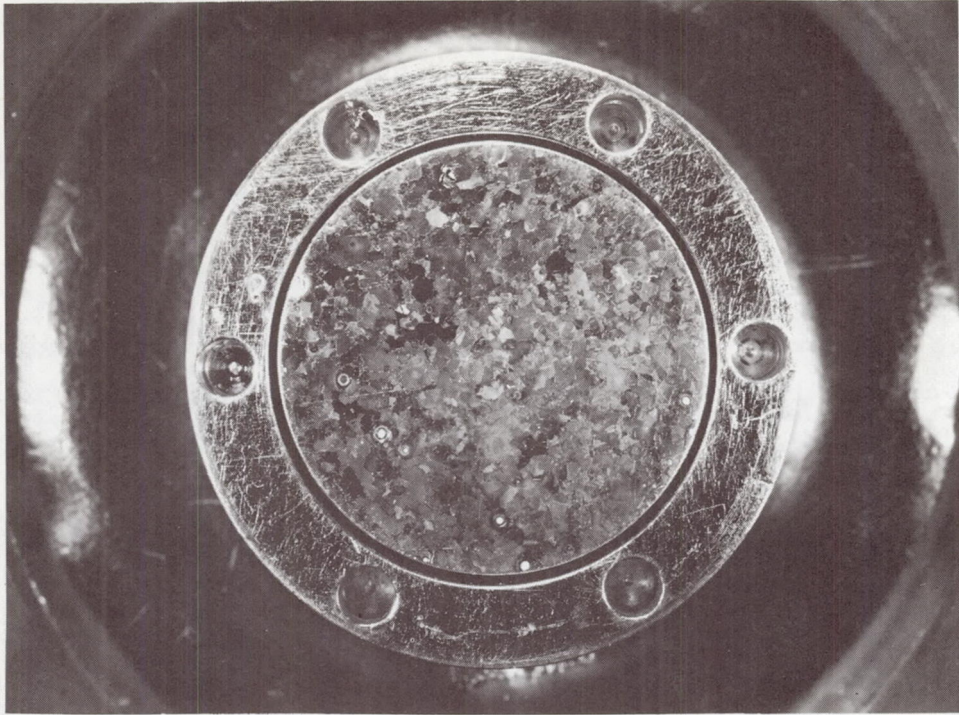
FIGURE 14a. COLLECTOR AND GUARD RING AFTER OPERATION
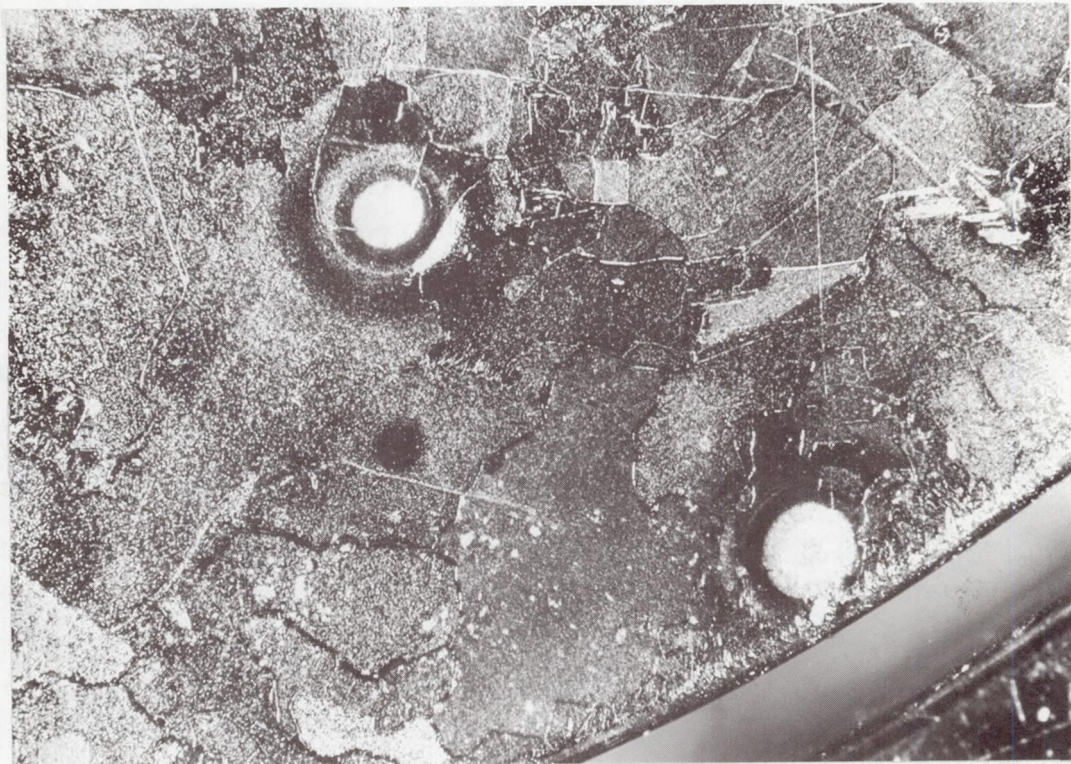(MAGNIFICATION – 4.2X)
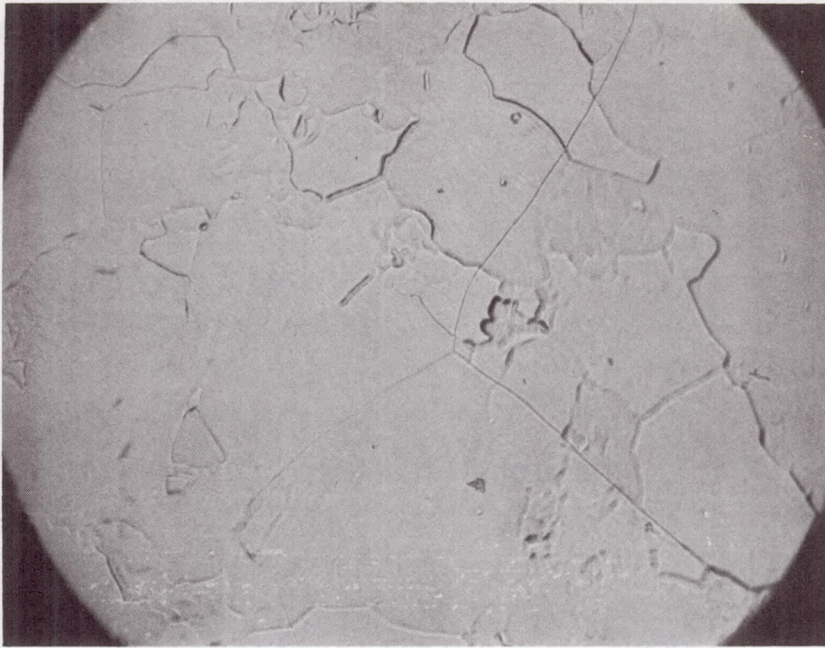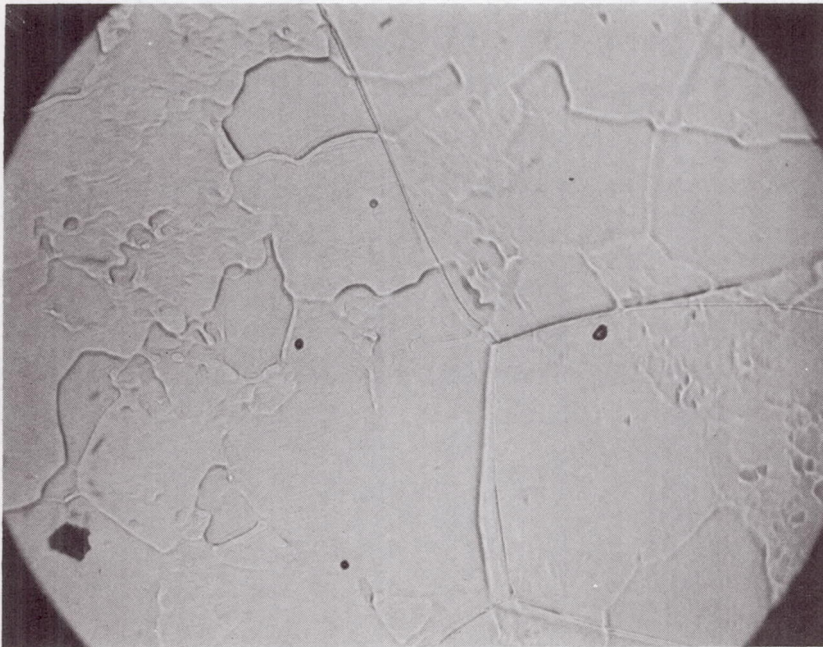

FIGURE 14b. ENLARGEMENT OF TWO "DISCHARGE" SPOTS ON COLLECTOR
(MAGNIFICATION – 42X)

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FIGURE 15a. EMITTER 1 BEFORE CONVERTER OPERATION — DIAMETER = 0.53 mm

FIGURE 15b. EMITTER 1 AFTER CONVERTER OPERATION — DIAMETER = 0.53 mm

not correct for the temperature difference between the back side and the front side of the tungsten emitter. They estimate that during the time they were taking the data, this ΔT was about $40^\circ K$. Figure 16 compares the output power of the Athanis-Van Someren converter with that of the converter reported here as a function of emitter temperature and current density. The open circles represent data presented by Athanis and Van Someren corrected for voltage and temperature as explained above. It is surprising that the two converters had almost identical output powers because the Athanis and Van Someren converter had a 2-mil spacing and a molybdenum collector whereas the converter of this report had a 5-mil spacing and a nickel collector. From tests on earlier converters, the increased output when going from 5-mil spacing to 2-mil spacing is usually not very great; so possibly a nickel collector is sufficiently better than a molybdenum collector to compensate for the difference in the spacing. The vapor deposited polycrystalline tungsten emitter oriented with the (110) crystallites exposed is as good as a single crystal (110) tungsten emitter.

Figure 16 shows for comparison, data from two other similar converters: (a) The converter by Howard, Van Someren and Yang had an identical emitter to that of this report, a molybdenum collector and a 5-mil spacing. Their preliminary report only gave one load line. The output at 10 amps/cm^2 is shown. (b) The converter reported by Kitrilakis and Rufeh had an electro-etched rhenium emitter with a work function of 4.88 eV, and a molybdenum collector. Complete families of load lines were given. For comparison, the points shown are for 20 amps/cm^2 and 5-mil spacing. As one might expect, the lower vacuum work function of their emitter results in slightly less power output.
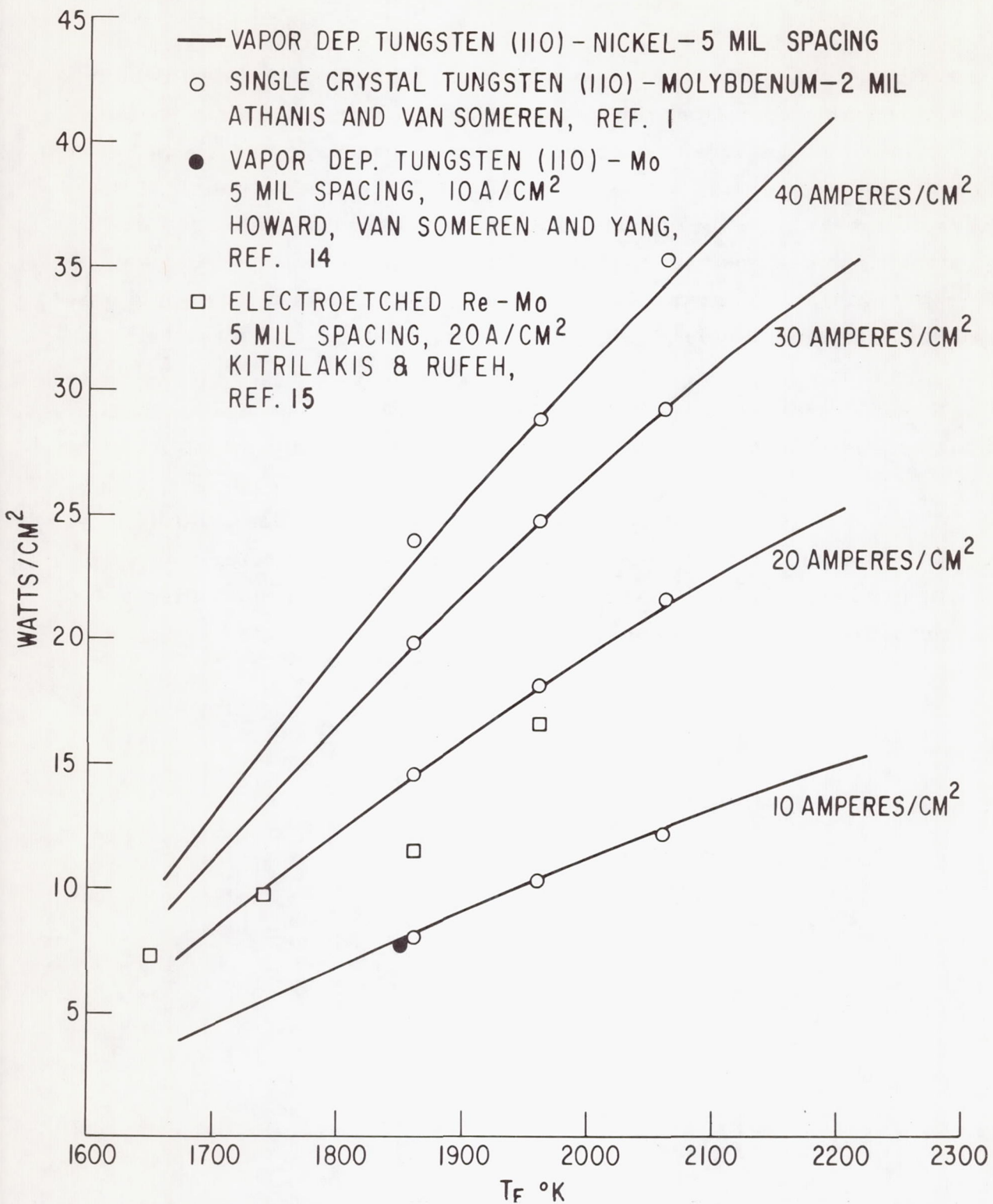
FIGURE 16.   COMPARISON OF A CONVERTER WITH A SINGLE CRYSTAL (110)
SURFACE WITH THE CONVERTER WITH THE POLYCRYSTAL VAPOR
DEPOSITED (110) SURFACE

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CONCLUSIONS

Although this converter was not operated for a long period of time to demonstrate stable operation with a long life, there were no indications of a reduction in output power with operation. Visual observation and work function measurements made during extensive emitter heat treatments indicated that the crystallites with the (110) planes exposed were the most stable and would grow at the expense of neighboring crystallites not so oriented. The sample used for the emitter was nearly 100% (110) oriented; therefore, one would expect it would be extremely stable at normal operating emitter temperatures.

The converter with the (110)-oriented polycrystalline tungsten emitter gave almost identical output as one with a single-crystal (110) tungsten emitter.

There was evidence of improvement in the nickel collector after standing in "vacuum" for one month. The "aged" nickel collector may have had a slightly lower cesiated work function than the molybdenum collectors of converters with which this converter was compared.

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