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

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

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16. Abstract

A parallel plane, variable spaced laboratory thermionic converter consisting of a chloride vapor deposited tungsten emitter of (110) crystal orientation and a collector of molybdenum surface layer deposited on a niobium base was built and tested. Current-voltage characteristics were obtained for a range of emitter temperatures from 1650 to 2035° K, collector temperatures from 993 to 1023° K, cesium reservoir temperatures from 573 to 653° K, and interelectrode spacings from 5 to 20 mils. Post-test analyses showed the collector surface to be molybdenum and niobium. The collector thermionic characteristics were primarily those of niobium.

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FOREWORD

The research described in this report was conducted by the General Electric Company under NASA contract NAS 3-8511. Robert P. Migra, of the Nuclear Systems Division, NASA Lewis Research Center, was the NASA Project Manager. The report was originally issued as General Electric report GESP-9011.

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ILLUSTRATIONS

SUMMARY

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

This sixth and last of a series of laboratory-type planar thermionic converters was of a new design. In this design, the envelope of the new converter was made entirely of niobium (Nb) except for two niobium-to-alumina insulating seals, the tungsten (W) emitter, and a tungsten-25 weight percent rhenium (W-25 w/o Re) foil supporting the emitter.

This converter had a chloride vapor deposited W emitter which had a preferred (110) crystal orientation on the surface as determined by x-ray diffraction. The collector was fabricated by depositing a 1500 Angstrom (Å) layer of molybdenum (Mo) on a Nb substrate.

The vacuum work function (ϕ_E) of the W emitter was measured prior to introducing cesium (Cs) into the converter. A value of 5.03 electron volts (eV) was obtained at an emitter temperature (T_E) of 2075 to 2300°K. After introducing the Cs but before operation of the converter, the collector work function (ϕ_C) was measured. A minimum ϕ_C of 1.54 eV was obtained at a ratio of collector temperature (T_C) to Cs temperature (T_{Cs}) of 1.9. The output performance of the converter was measured with emitter temperatures ranging from 1650 to 2035°K, Cs temperatures from 573 to 653°K, collector temperatures from 973 to 1023°K and interelectrode spacings from 59 to 20 mils. Early in testing, the collector and emitter were inadvertently shorted and the electrodes were separated while the

driving circuit was set to draw several hundred amperes. The resultant arc produced pits and molten balls on both the emitter and guard ring that prevented testing the converter at a spacing less than 5 mils.

The output performance of this converter was very similar to a converter that used a CVD-W emitter of (110) crystal orientation and a Nb collector. Post-test analyses of the collector surface revealed that it was Mo and Nb in about equal quantities. Thermionic performance indicated that the collector surface displayed characteristics of Nb.

INTRODUCTION

This work is part of a continuing program to build and operate thermionic converters with various electrode materials in order to characterize, evaluate, and identify the most promising electrode surfaces for converter operation. The design of the test converter was conceived and standardized in 1963. It permitted an accurate determination of the electrode spacing and used a guard ring to accurately define the converter area. The guard ring could be kept at the same temperature and potential as the collector. The first six lines of Table 1 list the electrode materials and the spacings for six converters built according to the 1963 design. The output power from these converters was consistently high and variations in output power could be explained by variations in electrode surfaces. The emitters of these converters were preheated to 2500°C for 1/2-hour and all other parts of the converter were heated at least 100°C hotter during processing than the operating temperature for each part.

In 1966, under NASA sponsorship, the program was altered in two respects: (1) A more elaborate converter was built so that the electrode spacing of each converter could be varied. (2) A much more intensive program to characterize the emitters was inaugurated. The NASA program, covered by Task III of Contract NAS 3-8511, began with the first variable spaced diode tested--namely, item 7 of Table 1. This report presents the emitter and collector preparation and the test results for the last variable spaced converter (item 12, Table 1). The test results are documented in the references listed in the table.

It is generally agreed by workers in the field that Nb is an inferior material for thermionic converter collectors. Holland, Kay, and Yates[11, 12] report that Mo is superior to Nb as a

Table 1.

	Emitter	Collector	Spacing (Inches)
(1)	Polycrystalline Tungsten[1]	Ni	0.005
(2)	Polycrystalline Rhenium[1]	Ni	0.005
(3)	Polycrystalline Rhenium[2, 3, 4]	Ni	0.002
(4)	Polycrystalline Tungsten[4, 5]	Ni	0.002
(5)	Polycrystalline Tungsten[5]	W	0.002
(6)	W-25 w/o Re[6]	Ni	0.005
(7)	Polycrystalline Tungsten[7, 8]	Nb	0.001 to 0.020*
(8)	Vapor Deposited (100) W, (110) Etch[9]	Nb	0.001 to 0.020
(9)	Vapor Deposited (110) W[10]	Ni	0.005
(10)	Vapor Deposited (110) W	Nb	0.002 to 0.020
(11)	Vapor Deposited (112) to (114) W	W + WO_2 on Nb	0.002 to 0.020
(12)	Vapor Deposited (110) W	Mo and Nb	0.005 to 0.020

*This converter did not have a guard ring.

collector material. Nb matches alumina in thermal expansion. Therefore, for a nuclear-heated cylindrical thermionic converter, it is desirable to use Nb insulated by alumina for the collector structure. An obvious solution is to use the Nb for the collector structure but coat it with a thin layer of Mo to enhance the converter performance.

The primary objective of this test was to determine the performance characteristics of a (110) oriented W emitter and a Mo collector. The collector was fabricated by depositing a 1500 Å thick Mo layer onto a Nb structure.

DESCRIPTION OF THE CONVERTER

The emitter for this converter was the same emitter as used in item 9 of Table 1.[10] It is a W disk 1-inch in diameter and 1/4-inch-thick coated on the lower surface with a 0.025-inch-thick layer of CVD W. After removing the emitter from the W (110)-Ni converter of reference 10, its orientation was rechecked by x-ray diffraction. The surface has large crystallites observable with the naked eye but appears to be almost 100% oriented with the (110) planes parallel to the bulk surface.

The collector and guard were ground and polished planar and coated with a 1500 Å layer of Mo by evaporation in vacuum. The thickness of the Mo layer was determined by simultaneously coating a blank glass specimen which was weighed before and after coating. The thickness was then calculated on the basis of the weight increase and the total area of the glass blank assuming a uniform deposition.

In mounting the converter in a vacuum bell jar, the guard is rigidly mounted on three stainless steel rods. The collector is forced by springs against the ceramic ring insulating the guard from the collector. This ring determines the relative height of the collector and guard. Three movable stainless steel legs are clamped to the flanges on the large upper Nb piece referred to as the emitter radiator. The clamps contain sapphire balls that insulate the emitter radiator from the guard and bell jar base plate. For a more detailed description of the spacing mechanism, see NASA CR-1033 (pp. 8-11).[8]

Converters used in the previous tests often developed vacuum leaks during operation. Most of these leaks developed

in brazed joints between Ni and Mo parts. A new testing vehicle was designed which uses Al_2O_3 to Nb insulating seals. The emitter support and electrical lead is a W-25 w/o Re cylinder with a 0.005-inch-thick wall. The rest of the assembly is Nb. Except for the ceramic-to-metal seals, the entire assembly is joined by electron beam welds. Figure 1 is a cross section drawing of the assembly.

The ceramic pieces for the seals are high purity Al_2O_3 washers. The planar surfaces were metallized by a process developed by R. H. Bristow.[13] Briefly, a mixture of finely powdered calcia, magnesia, and Al_2O_3 was painted and fired onto the Al_2O_3 surface. The resulting surface was electroplated with a Cu coating. The seal assembly was made by stacking the Nb spinnings and metallized Al_2O_3 washers according to Figure 1 with 1-mil foils of Cu-37 w/o Au (Melting Point of 1025°C) placed between the Nb and metallized ceramic surfaces. The assembly was held under compression and fired to 1050°C. The Cu-Au foils were made slightly smaller than the ceramic rings to prevent the braze from running down the cylindrical surfaces of the ceramics. Cu-37 w/o Au was used because Cu-Nb forms a liquidus phase at 1125°C and it is difficult to make a Cu braze just a few degrees lower without dissolving part of the Nb foils.

The 5-mil-thick, 1-inch-long W-25 w/o Re cylinder was made by rolling a foil and seam welding a butt joint. This cylinder was welded to the W emitter and then to the large radiator section. Therefore, three practice welds were made between the W-25 w/o Re foil and Nb and heat cycled 25 times each from room temperature to 1000°C. They remained vacuum tight. However, after the first converter of this design was assembled, cracks developed in the W-Re foil below the weld to Nb. A second emitter and W-25 w/o Re foil assembly was annealed to 1200°C in wet hydrogen (H_2) and then to 1600°C in dry H_2. This foil remained vacuum tight after welding to Nb.

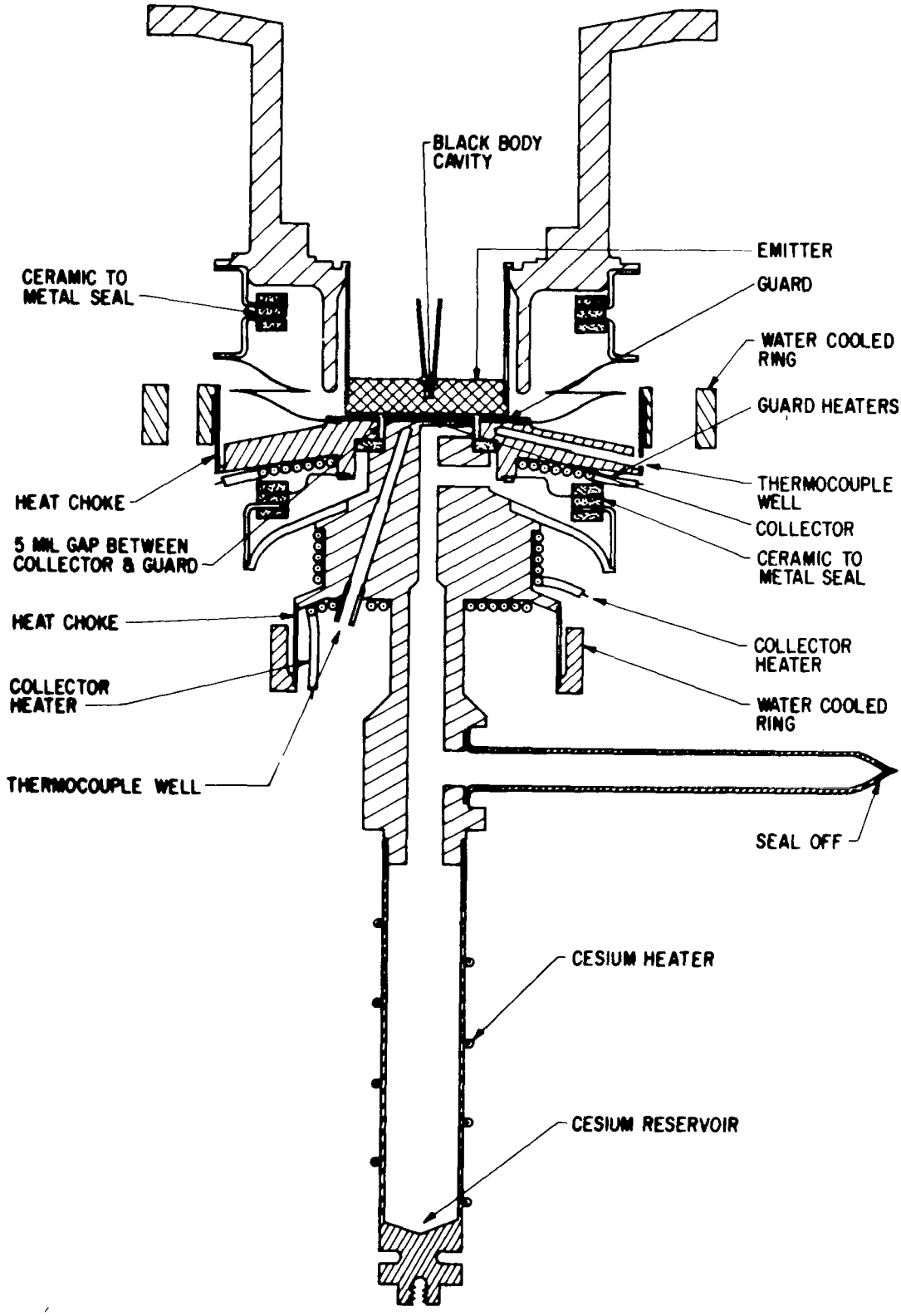

Figure 1. CROSS SECTION OF Nb, ALL WELDED, PLANAR CONVERTER

CONVERTER OPERATION

Before introducing the Cs, the work function of the emitter was measured. It remained constant at 5.03 eV from 2075°K to 2300°K.

After introducing the Cs but before operation as a converter, the work function of the collector was measured by saturation emission with the collector at 800 or 820°K and varying the Cs reservoir temperature from 409 to 493°K. Figure 2 shows a plot of ϕ versus T_C/T_{Cs} ϕ_{min} is 1.536 eV at $T_C/T_{Cs} = 1.9$.

The output performance of the converter was measured with T_E from 1650 to 2035°K, T_{Cs} from 573 to 653°K, T_C from 973 to 1023°K and spacings from 5 to 20 mils. The individual J-V curves obtained for these parameters are presented in Appendix A. Early in testing this converter, the collector and emitter were inadvertently shorted. The electrodes were separated while the driving circuit was set to draw several hundred amperes. The resultant arc produced pits and balling on both the emitter and guard. The balling prevented testing the converter at a spacing less than 5 mils.

The highest spot on the guard was 3.5 mils above the surface. The point on the emitter was 1.2 mils high. After the arc, the converter was releveled so that as it was brought together, the leveling legs were equally sensitive in producing short circuits. Therefore, the data were taken with the emitter tipped relative to the collector. A spacing reported as 5 mils probably varied from 4 to 6 mils from one side to the other of the collector.

The output performance appeared very similar to converter #10 in Table 1 which had a similar emitter but a Nb collector.

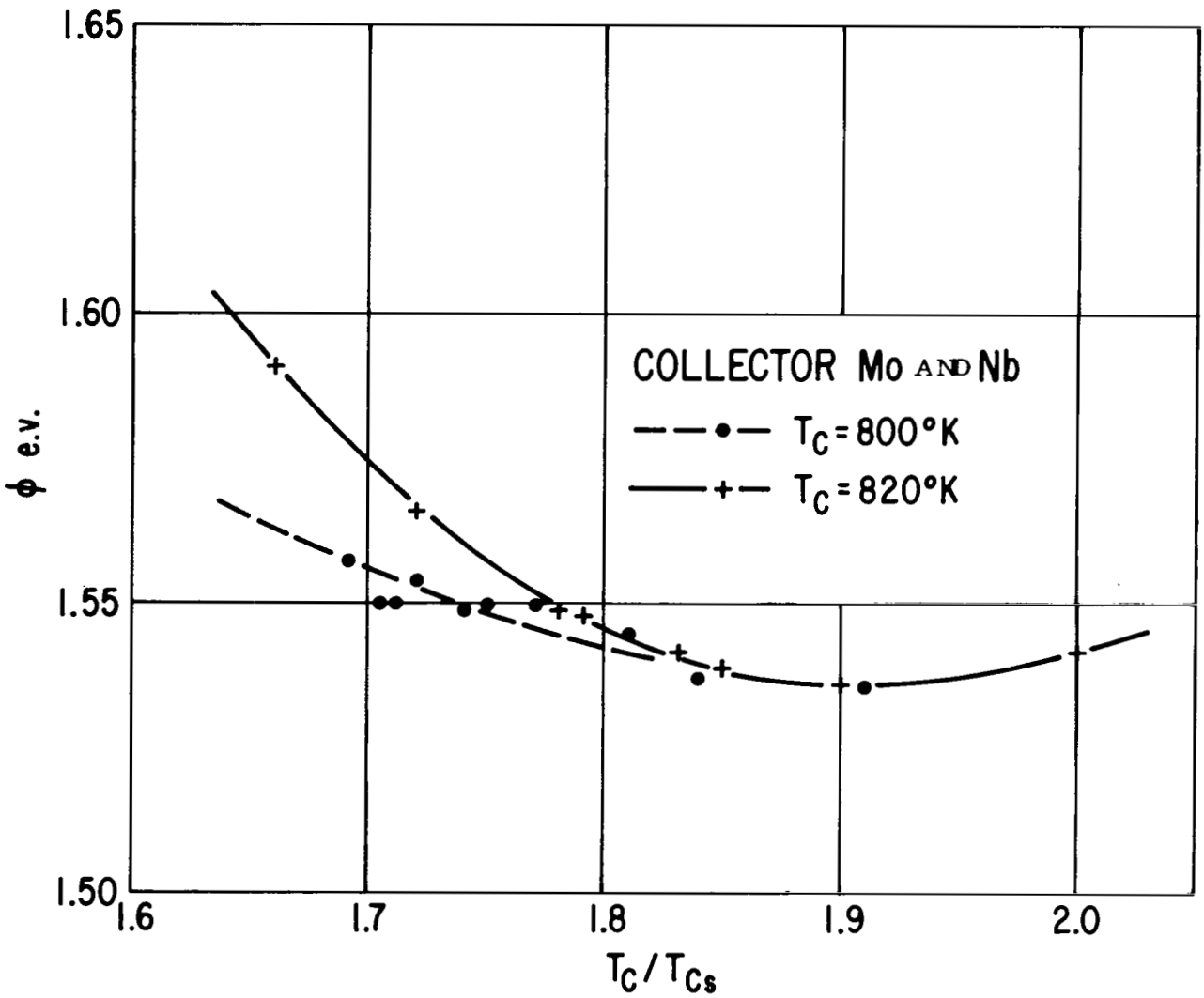

Figure 2. COLLECTOR WORK FUNCTION ϕ VERSUS T_C/T_{Cs} RATIO

This similarity is illustrated in Figures 3 and 4. These data indicate that the collector surfaces of the two converters are essentially the same, assuming that there are no differences in the emitter characteristics.

Because this converter with the Mo on Nb collector had the same output as a converter with a Nb collector and it was anticipated that the Mo on the collector would improve the output performance, it was suspected that Nb had diffused to the surface during the outgassing of the guard and collector which took a few hours at 900°C. The converter was taken apart and the collector surface was analyzed with an Auger electron spectrometer. The results revealed that the surface was Mo and Nb in about equal quantities. Data from N. L. Peterson[14] and Birks and Seebold[15] for diffusion of Mo and bulk Nb indicates that the Nb should not have diffused to the surface so quickly; however, recent measurements by J. R. Young of diffusion of Nb into a Mo evaporated layer give a diffusion coefficient larger by a factor of 5. Perhaps the evaporated Mo layer is not densely packed and Nb diffuses more rapidly through it than was anticipated.

The thermionic performance of a diode is highly temperature dependent. It is, therefore, important that the emitter temperature be well known. A calculation of the emitter thermal gradients occurring in these experimental converters is included as Appendix B.

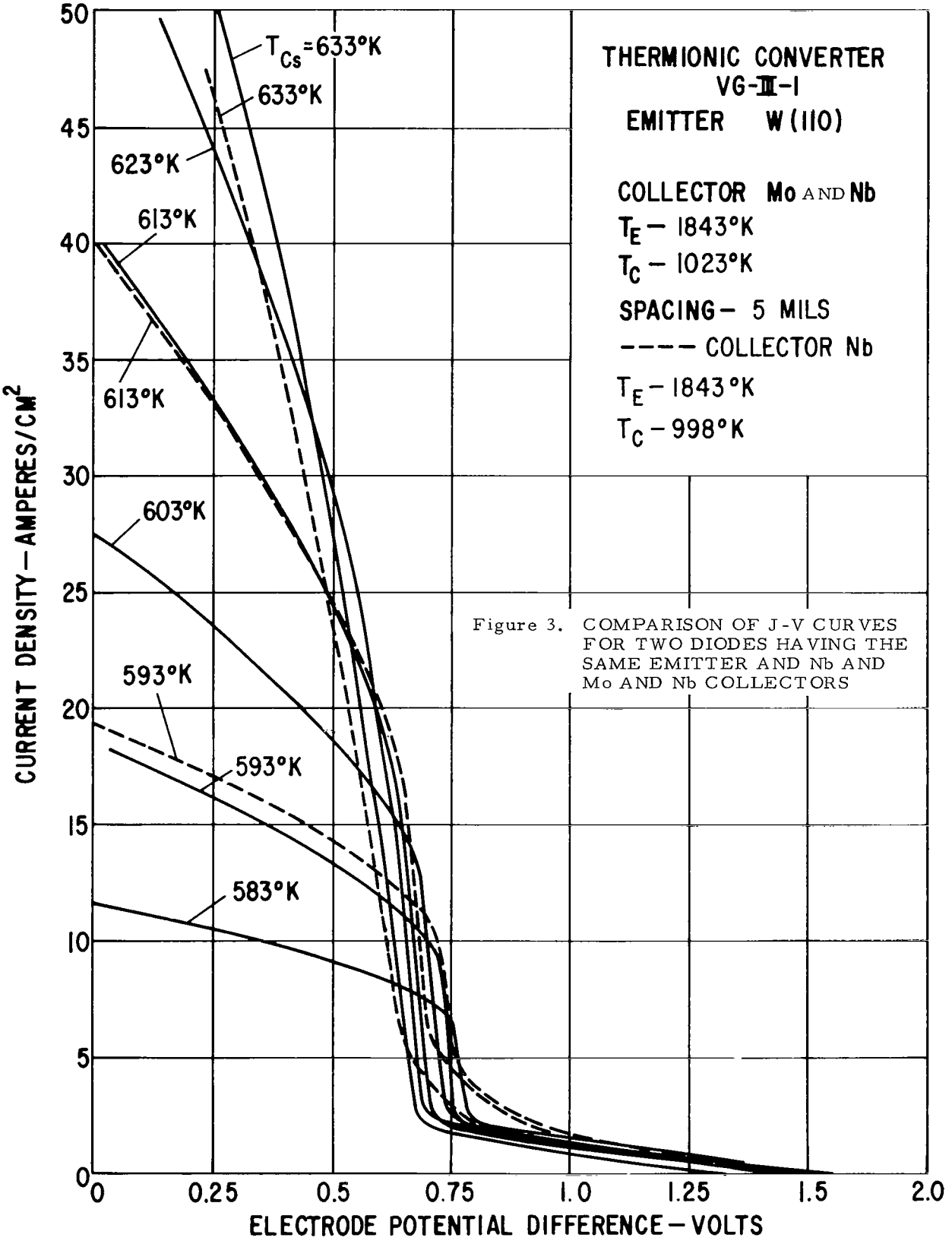

Figure 3. COMPARISON OF J-V CURVES FOR TWO DIODES HAVING THE SAME EMITTER AND Nb AND Mo AND Nb COLLECTORS

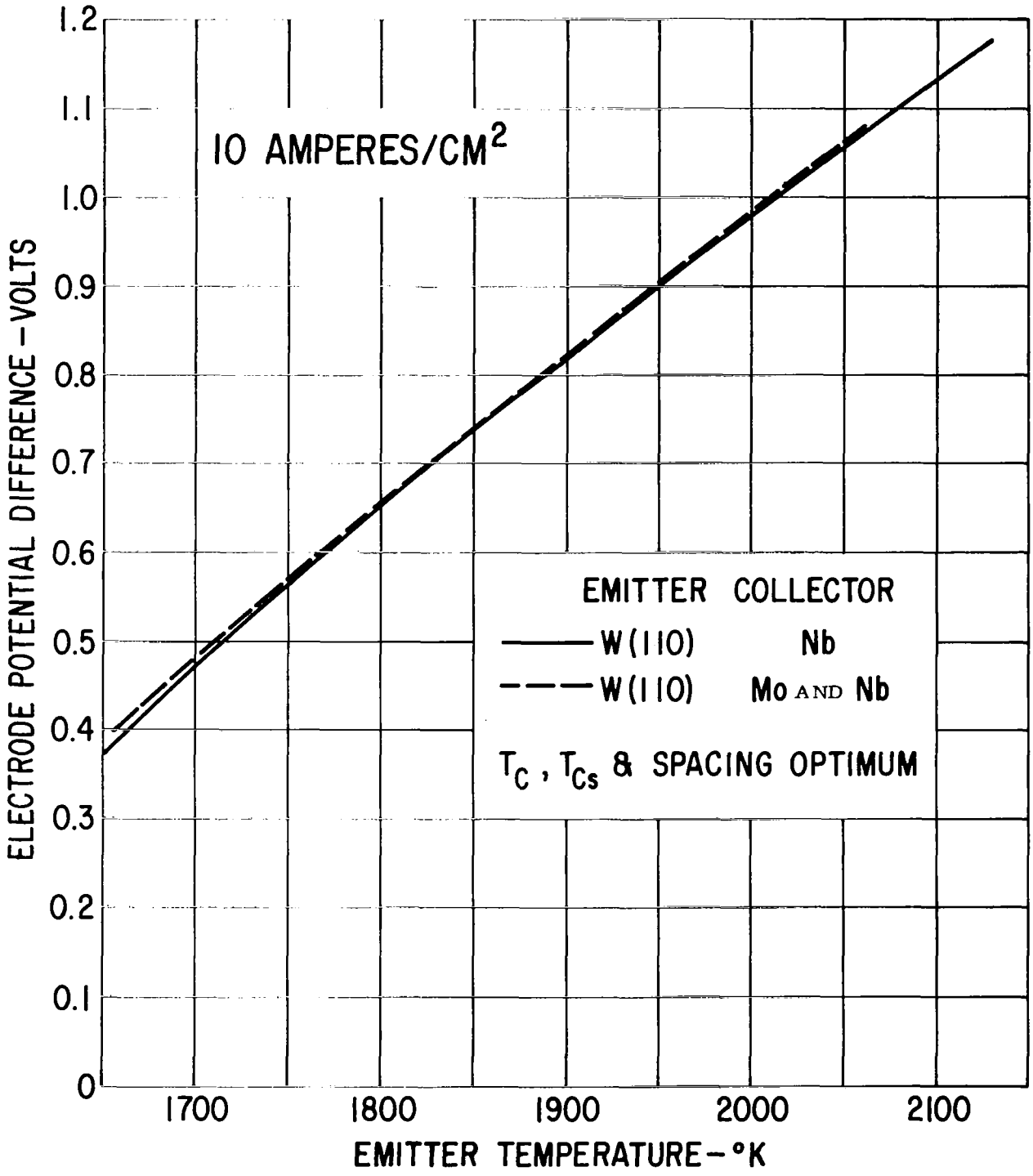

Figure 4. COMPARISON OF POTENTIAL DIFFERENCE AT 10 AMPS/cm^2 FOR TWO DIODES HAVING THE SAME EMITTER AND Nb AND Mo AND Nb COLLECTORS

CONCLUSIONS

There are two conclusions resulting from this work: (1) the performance results are for a mixed Mo and Nb collector surface, and (2) more work is required to determine a satisfactory method of applying Mo to a Nb structure for use as a collector for a long-life converter.

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APPENDIX A

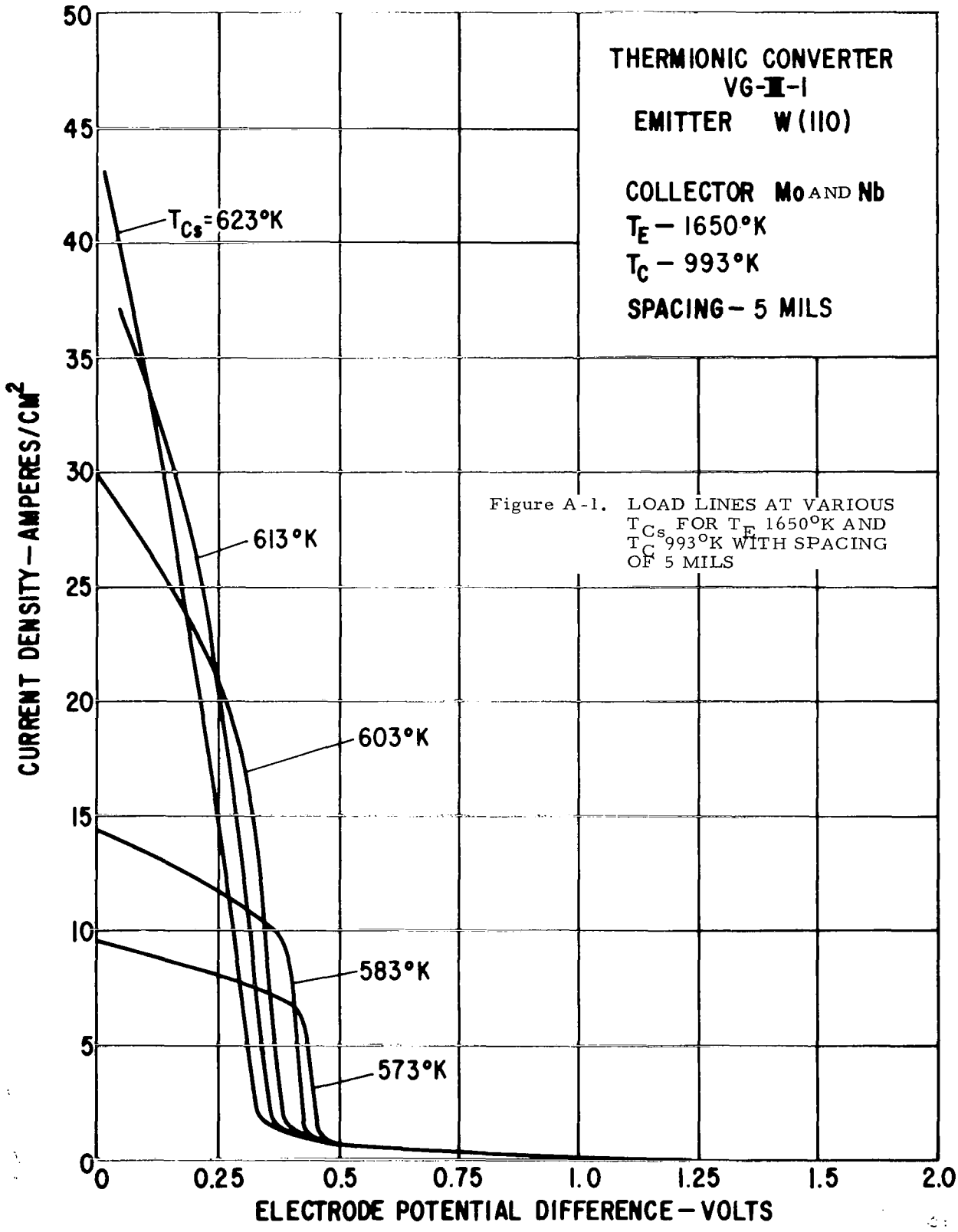

Figure A-1. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1650°K AND T_C 993°K WITH SPACING OF 5 MILS

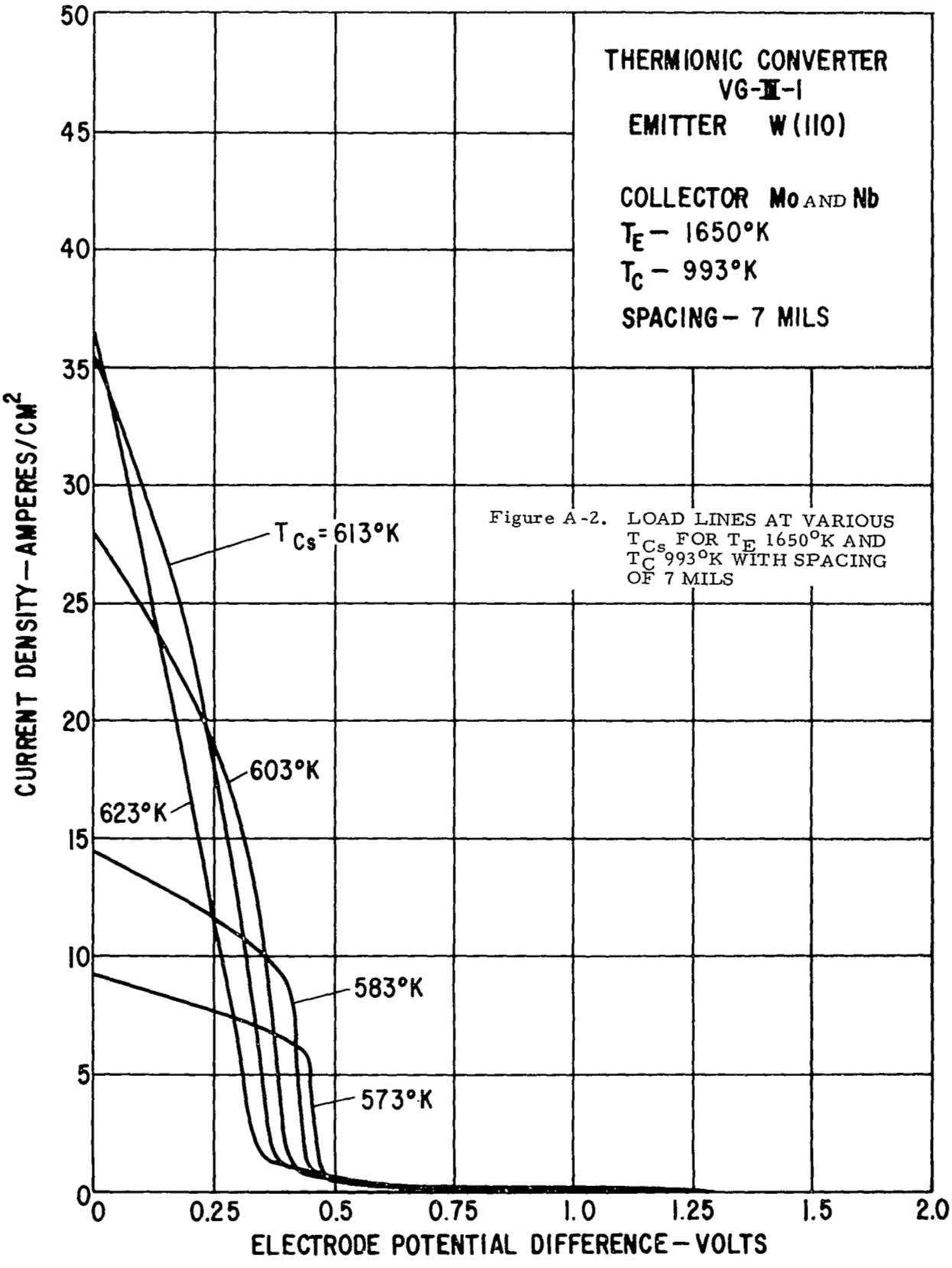

Figure A-2. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1650°K AND T_C 993°K WITH SPACING OF 7 MILS

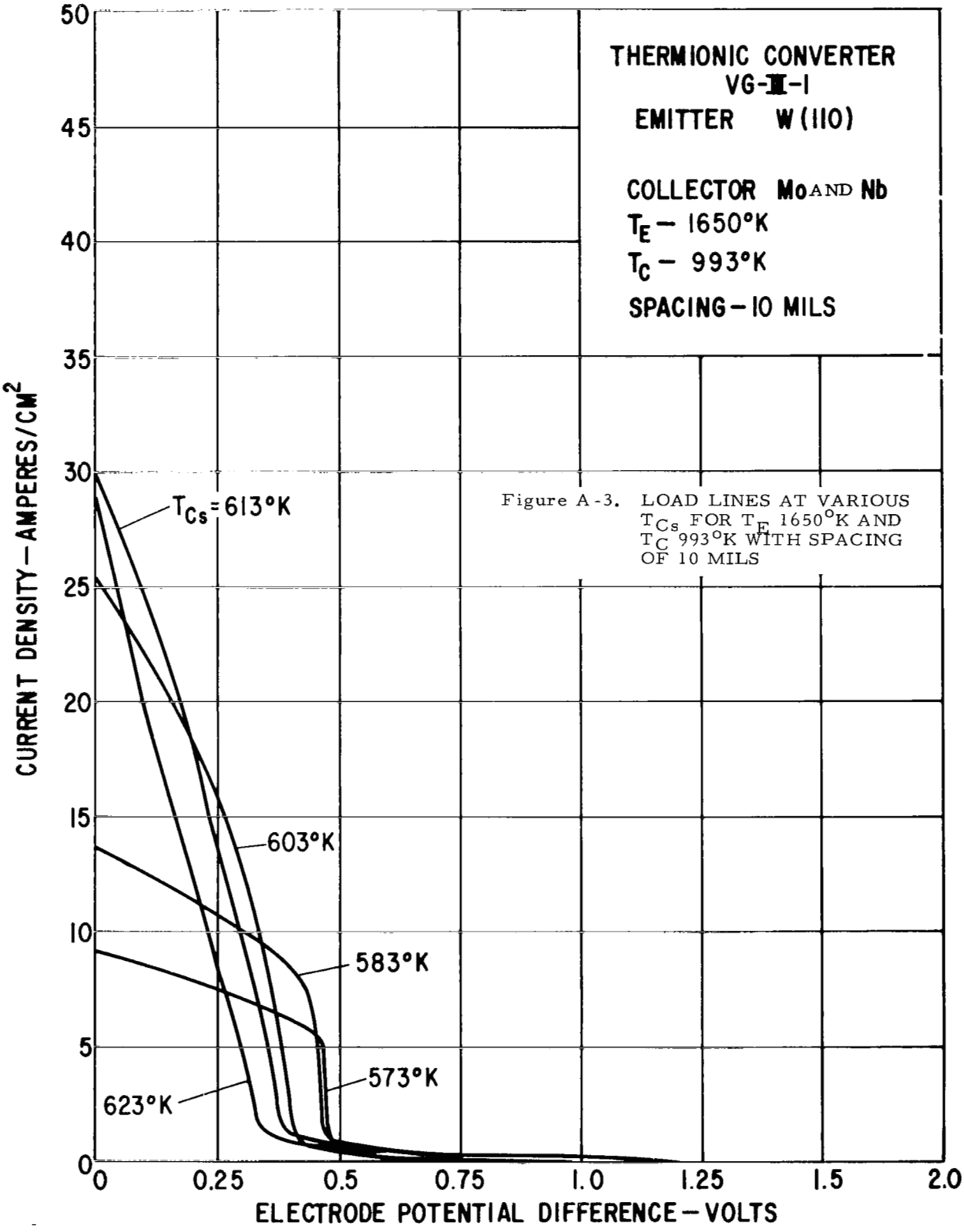

Figure A-3. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1650°K AND T_C 993°K WITH SPACING OF 10 MILS

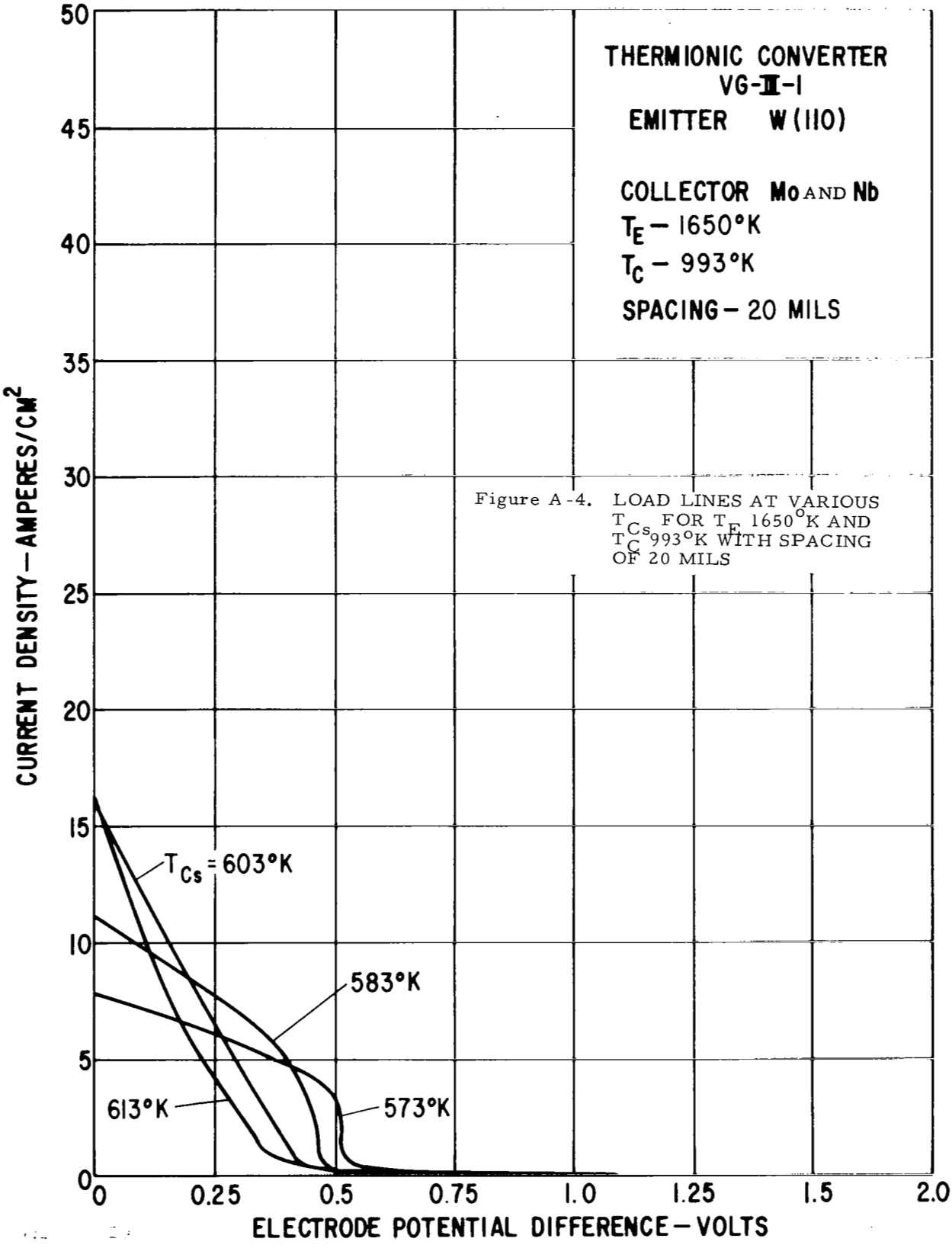

Figure A-4. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1650°K AND T_C 993°K WITH SPACING OF 20 MILS

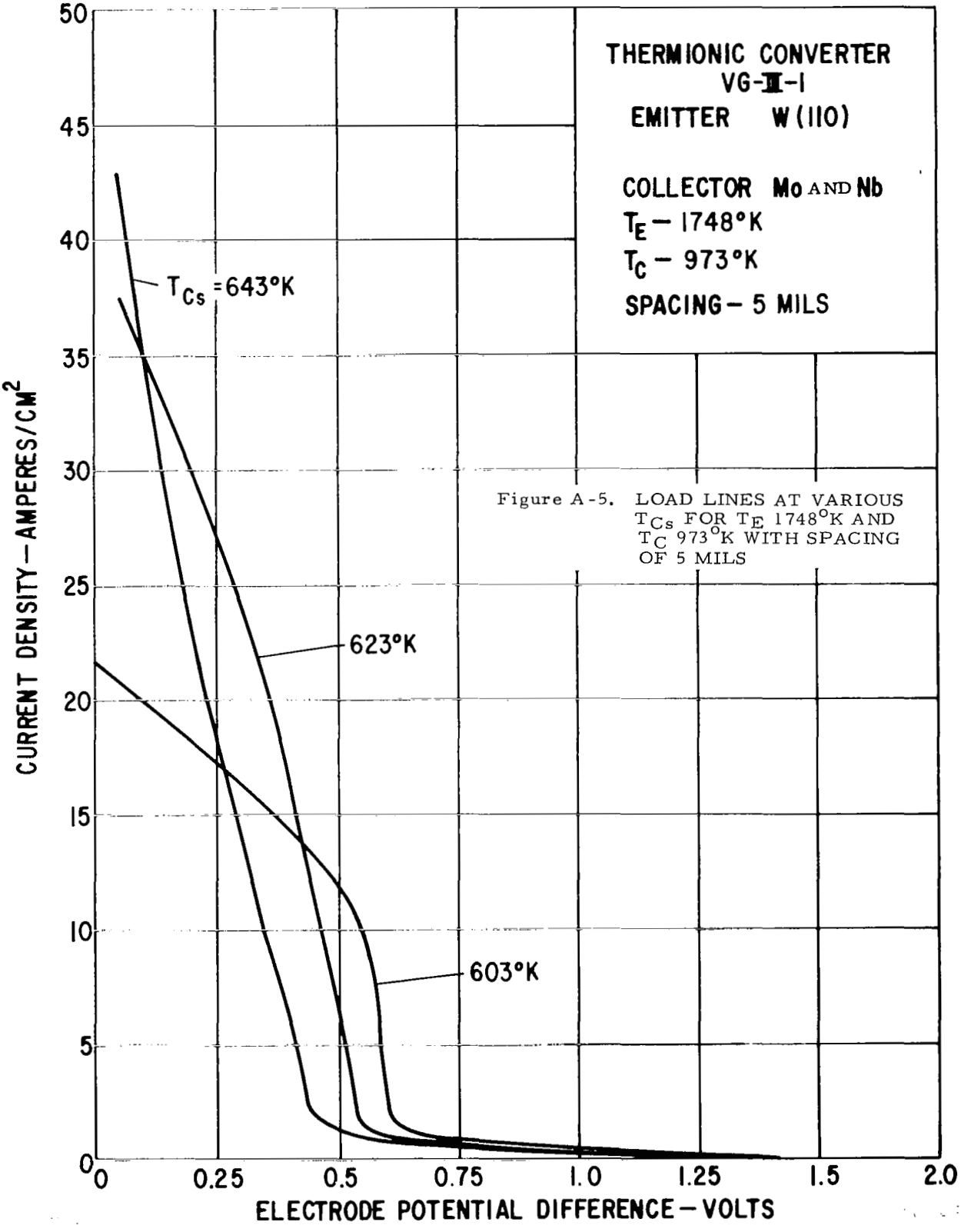

Figure A-5. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1748°K AND T_C 973°K WITH SPACING OF 5 MILS

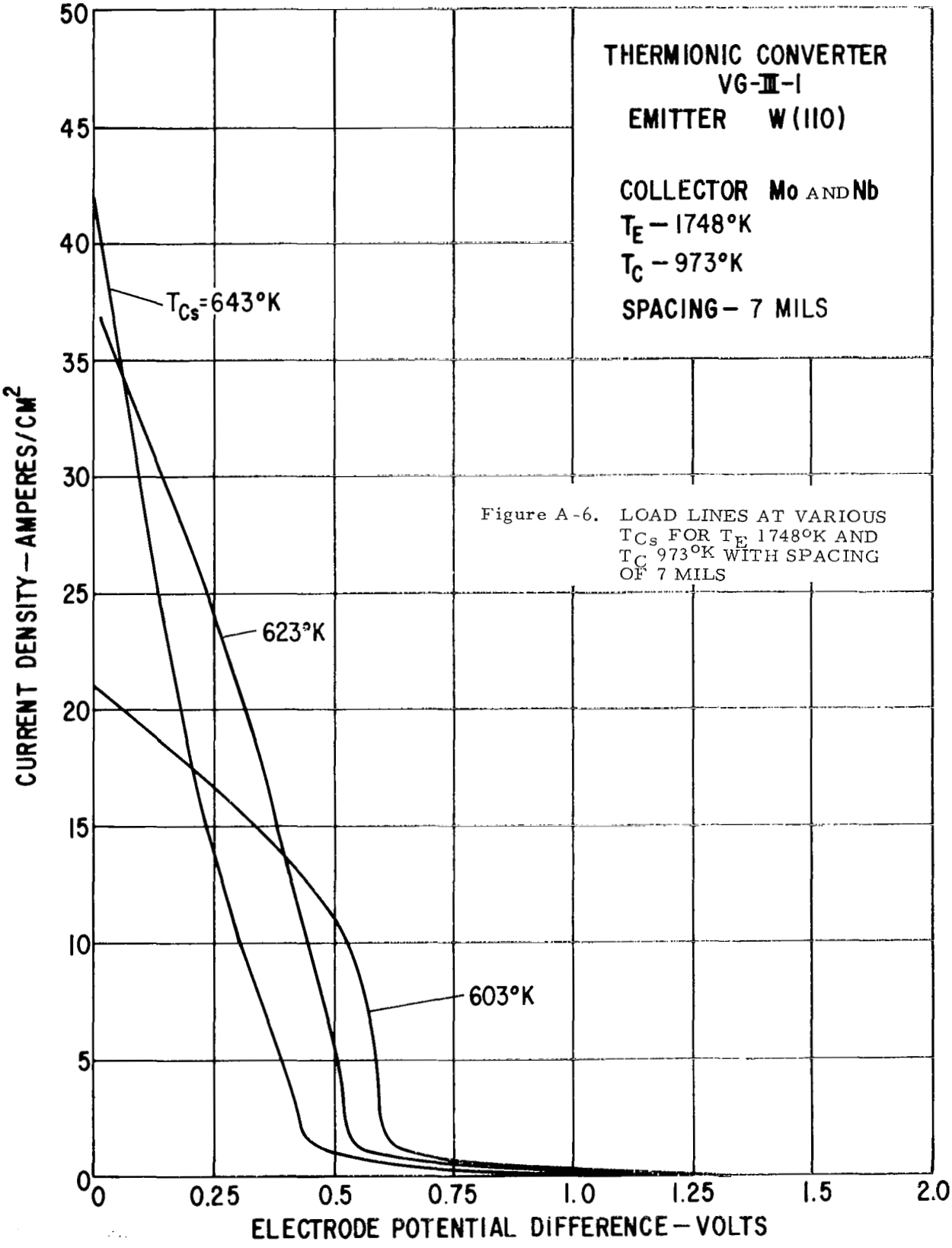

Figure A-6. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1748°K AND T_C 973°K WITH SPACING OF 7 MILS

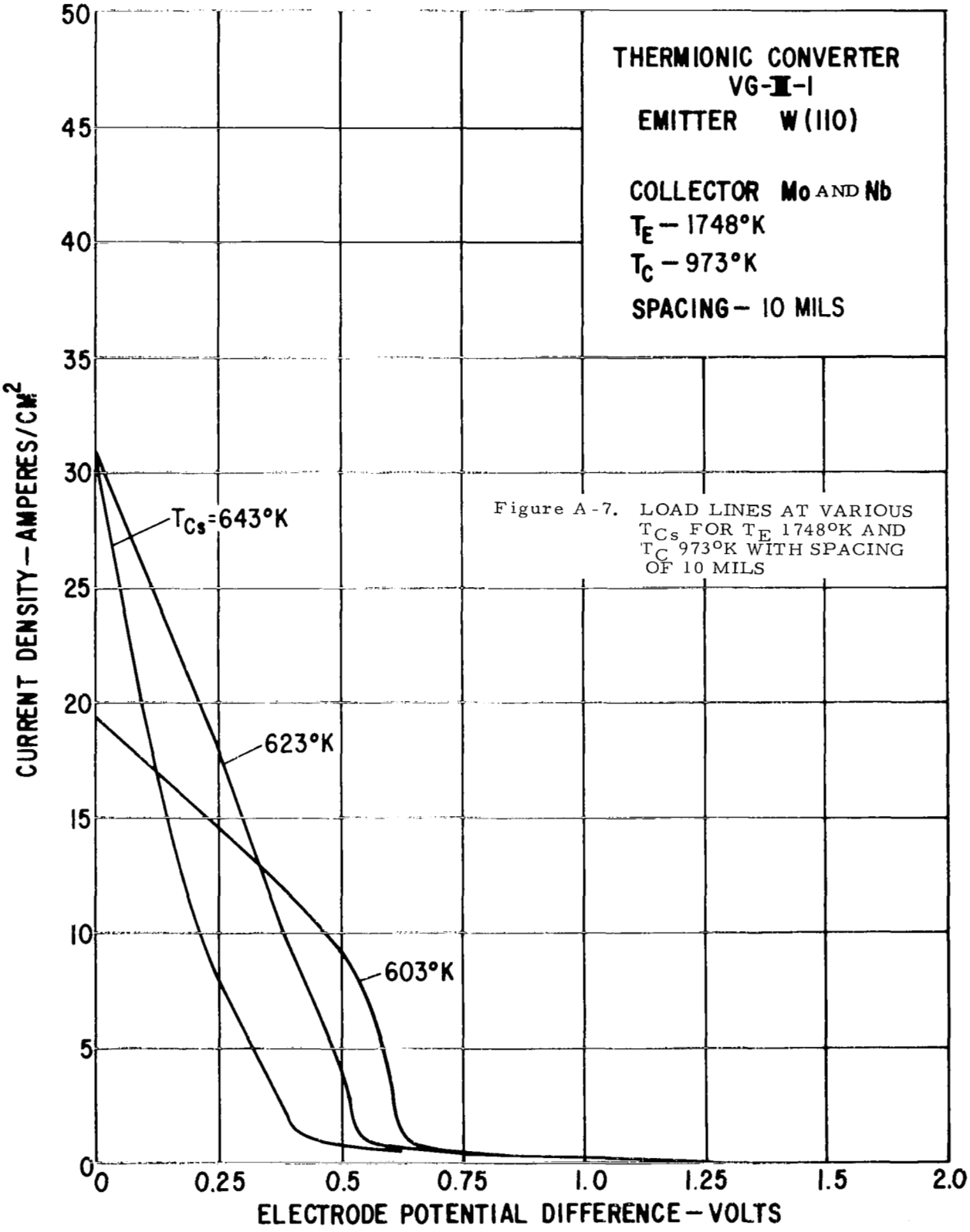

Figure A-7. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1748°K AND T_C 973°K WITH SPACING OF 10 MILS

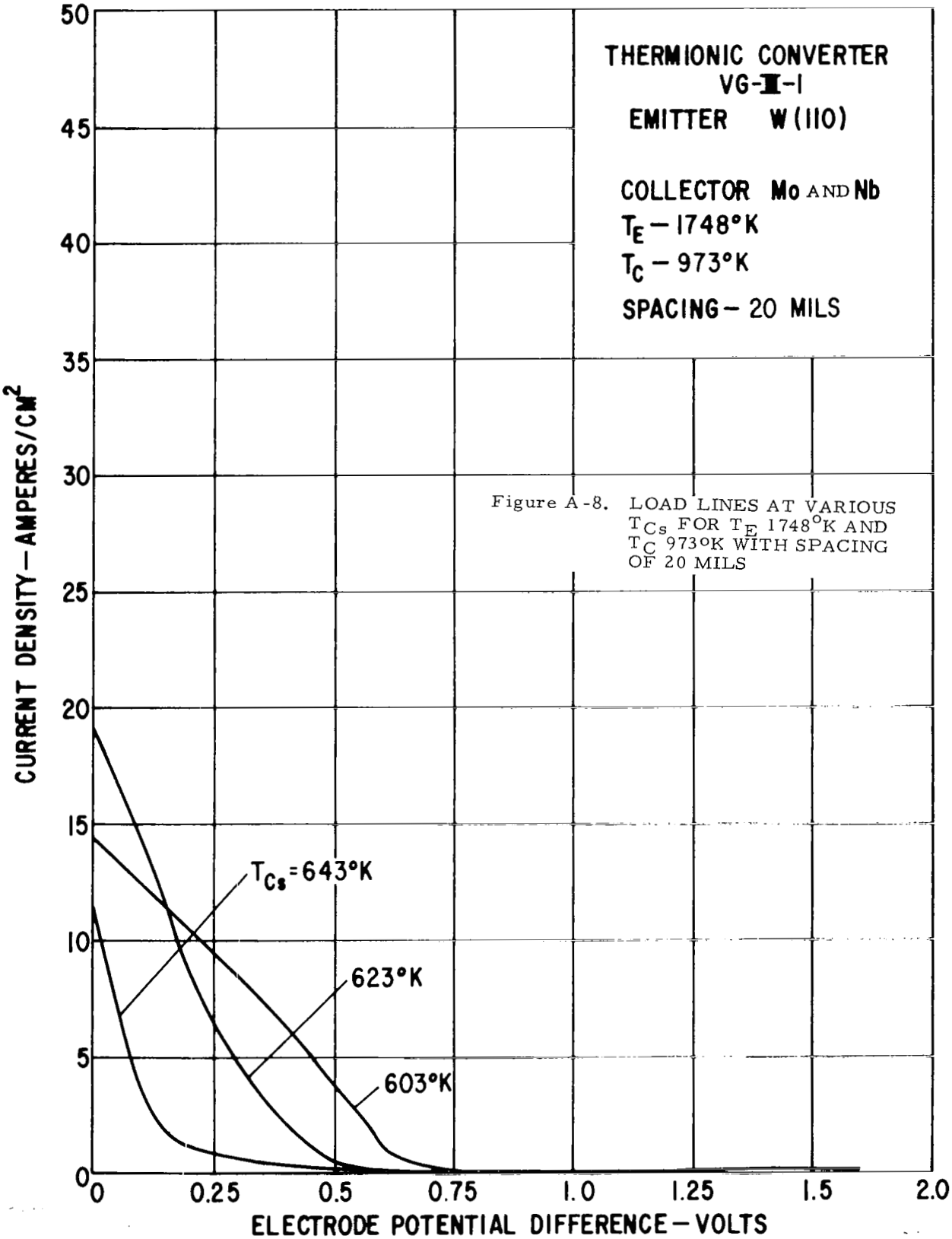

Figure A-8. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1748°K AND T_C 973°K WITH SPACING OF 20 MILS

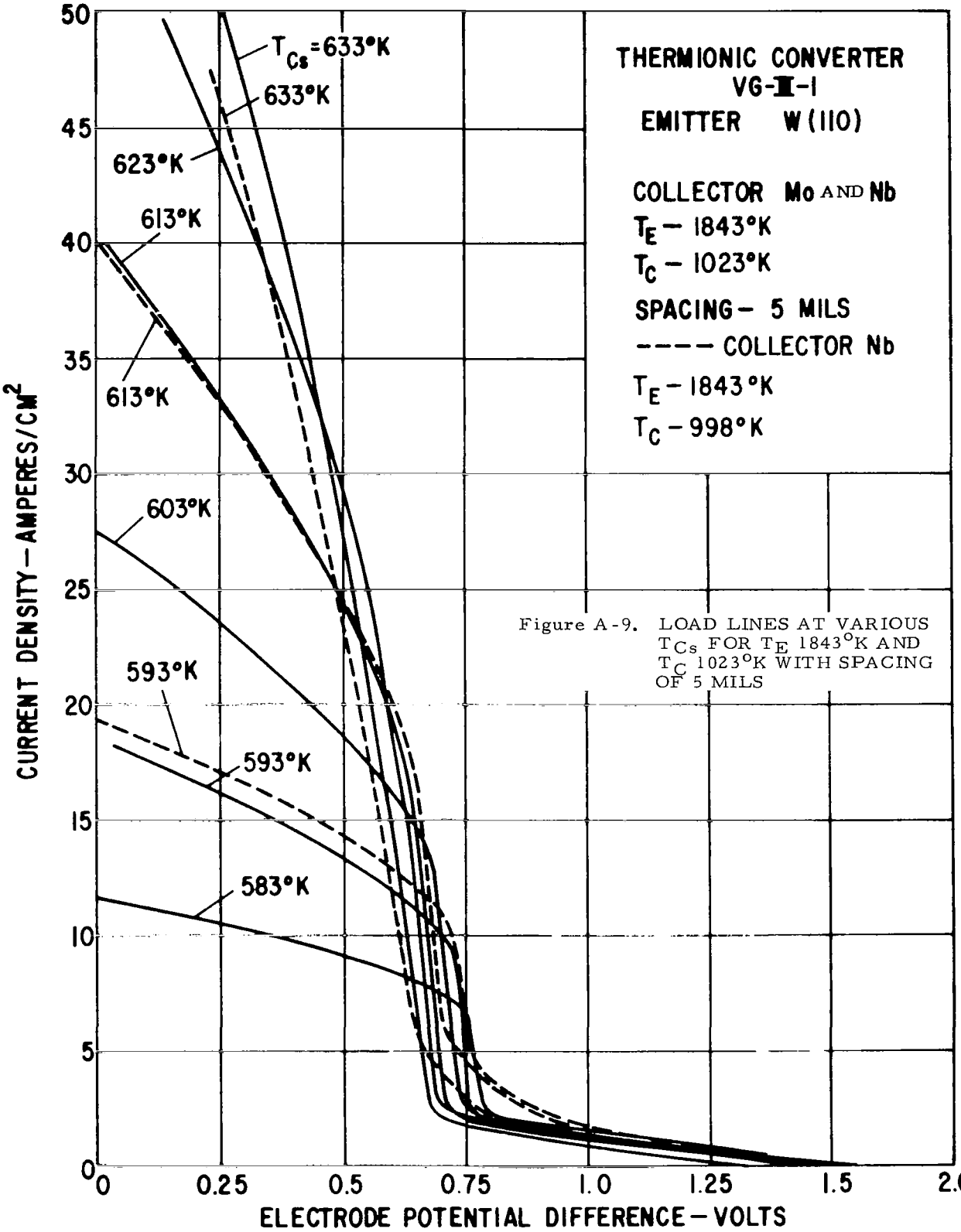

Figure A-9. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1843°K AND T_C 1023°K WITH SPACING OF 5 MILS

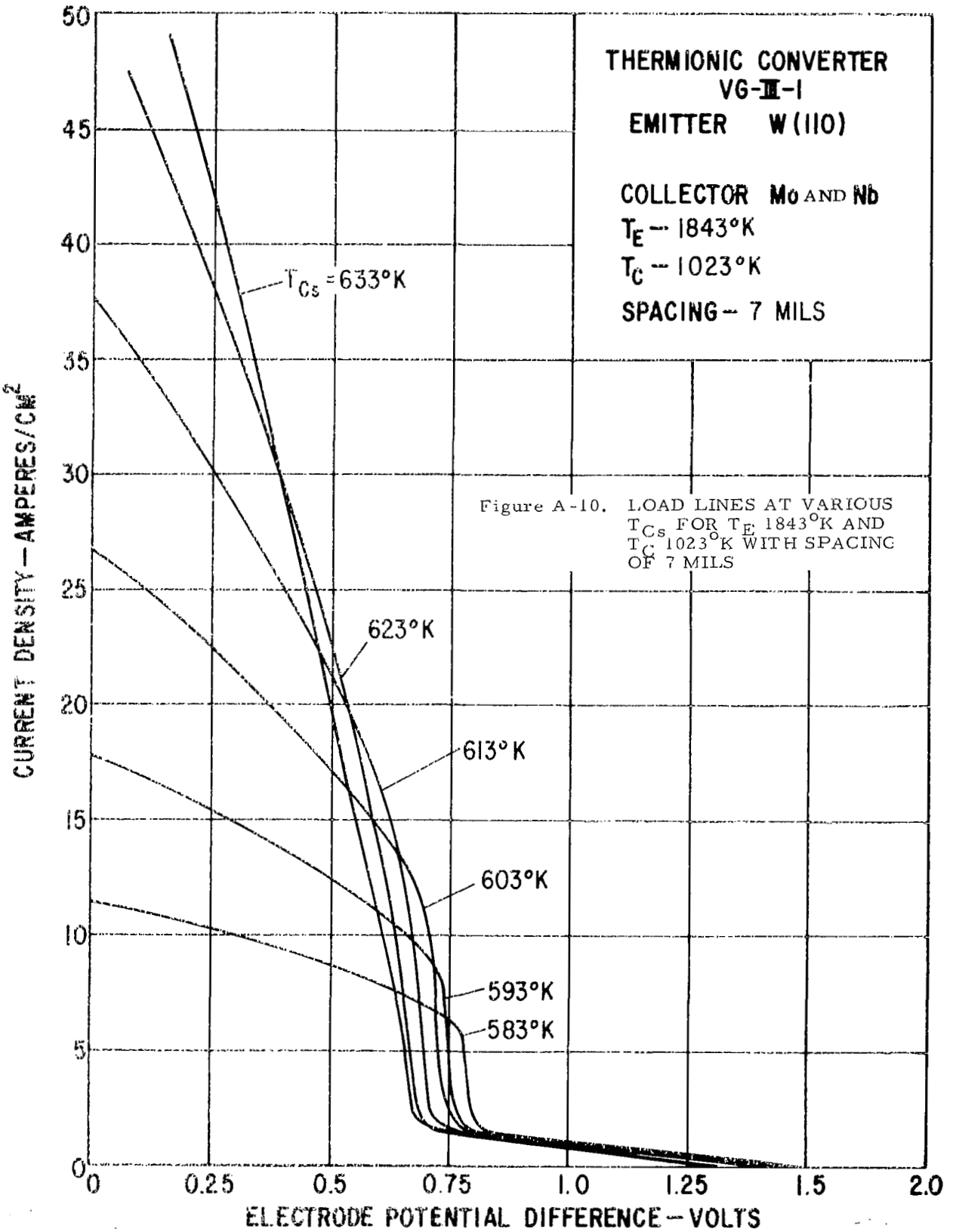

Figure A-10. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1843°K AND T_C 1023°K WITH SPACING OF 7 MILS

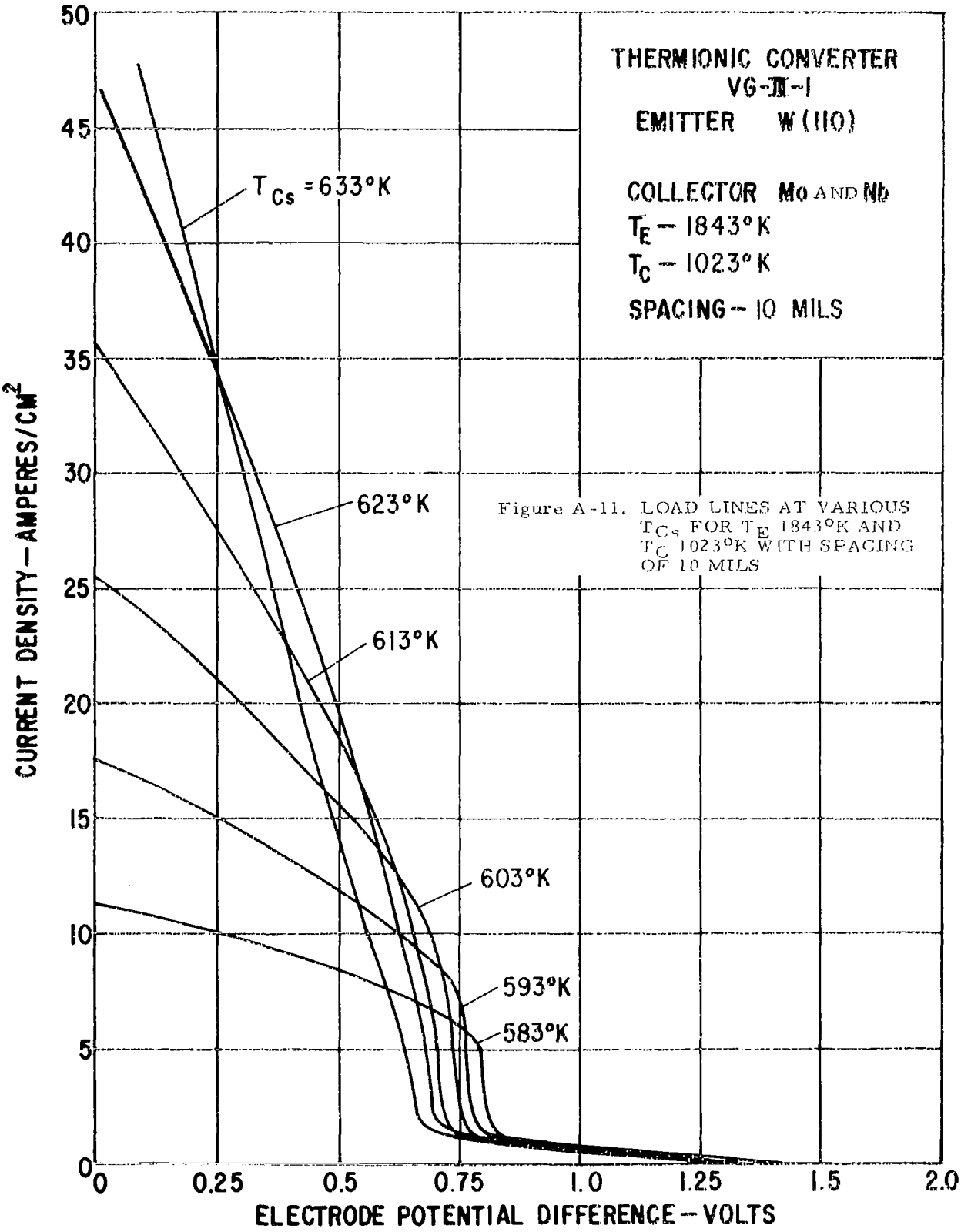

Figure A-11. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1843°K AND T_C 1023°K WITH SPACING OF 10 MILS

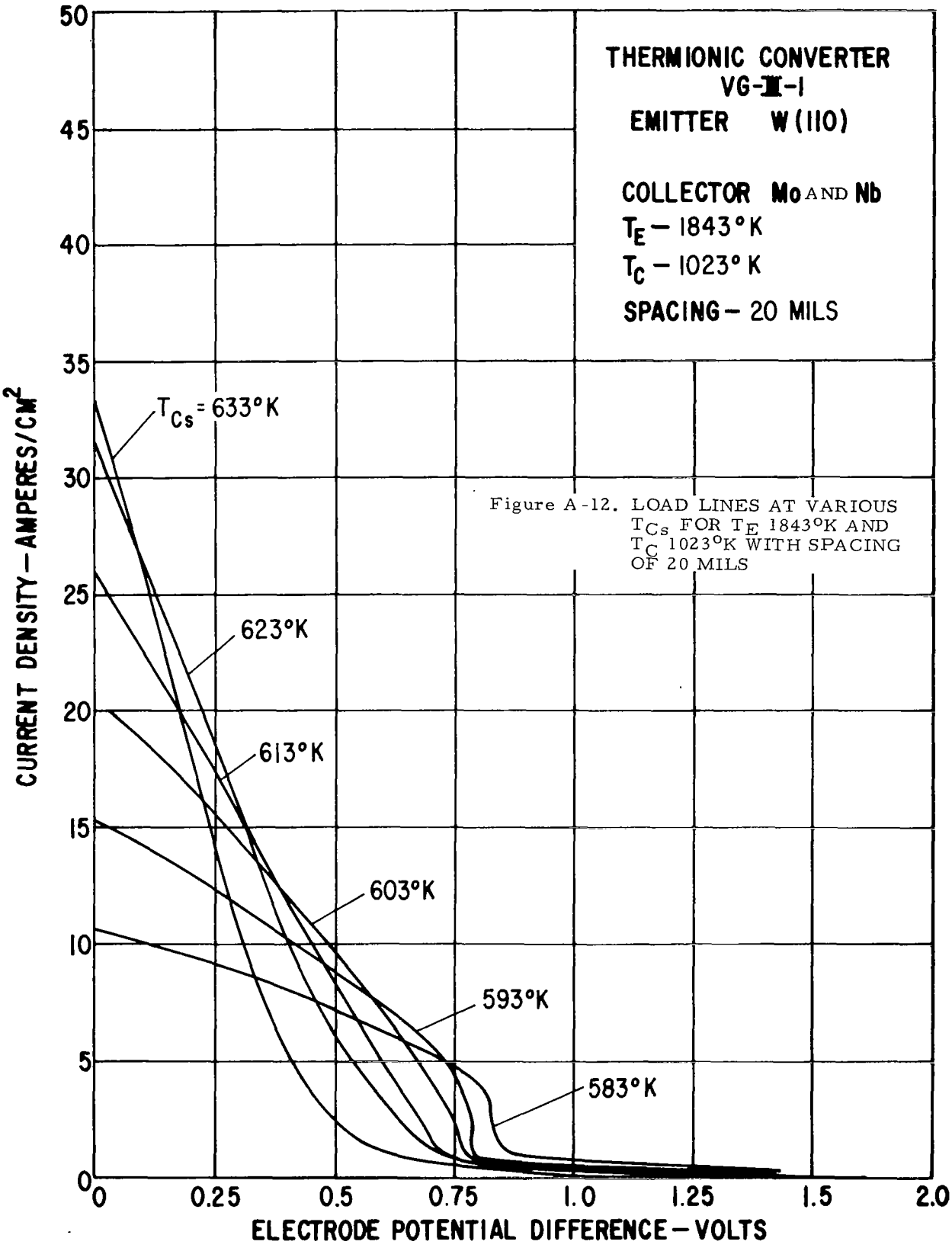

Figure A-12. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1843°K AND T_C 1023°K WITH SPACING OF 20 MILS

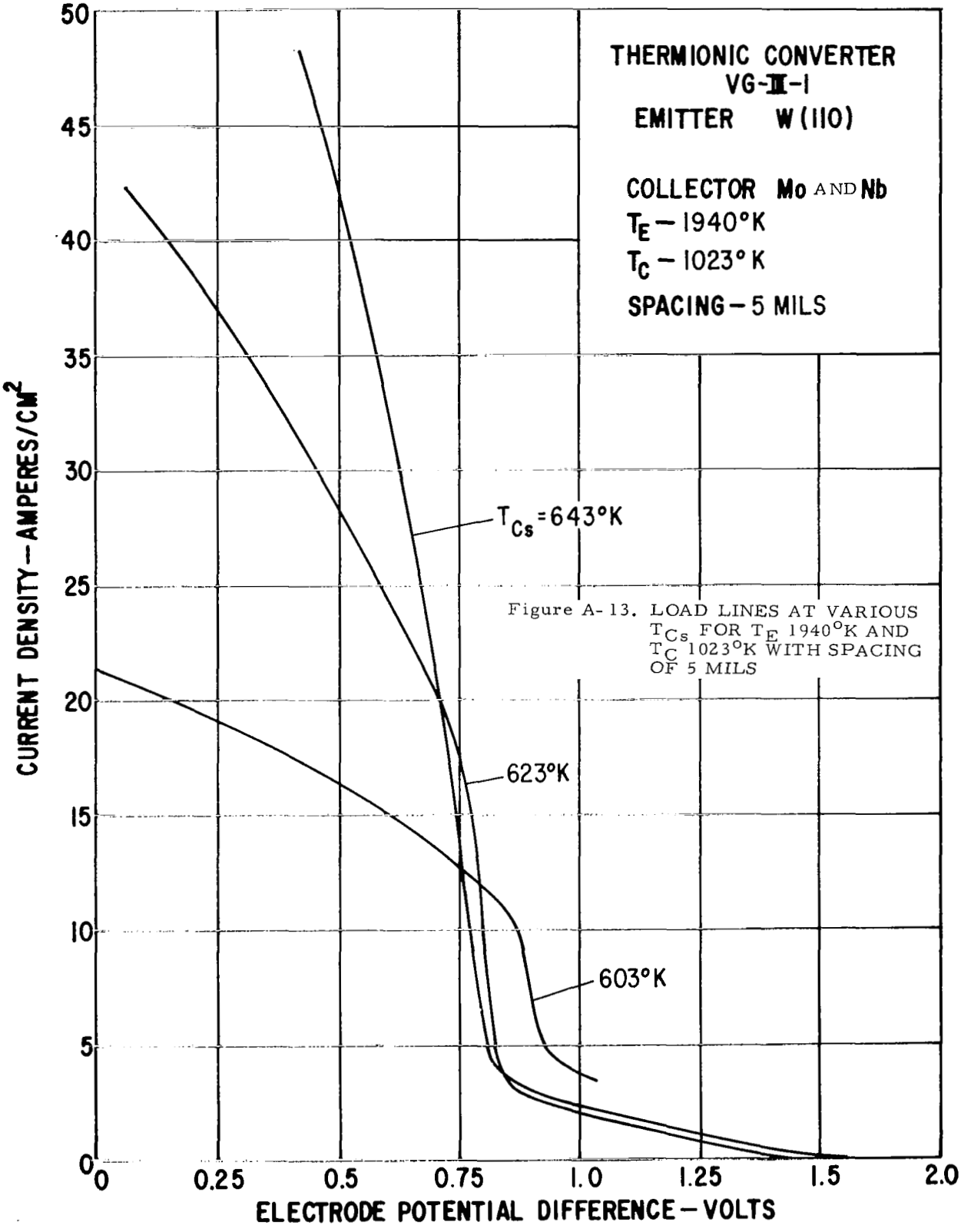

Figure A-13. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1940°K AND T_C 1023°K WITH SPACING OF 5 MILS

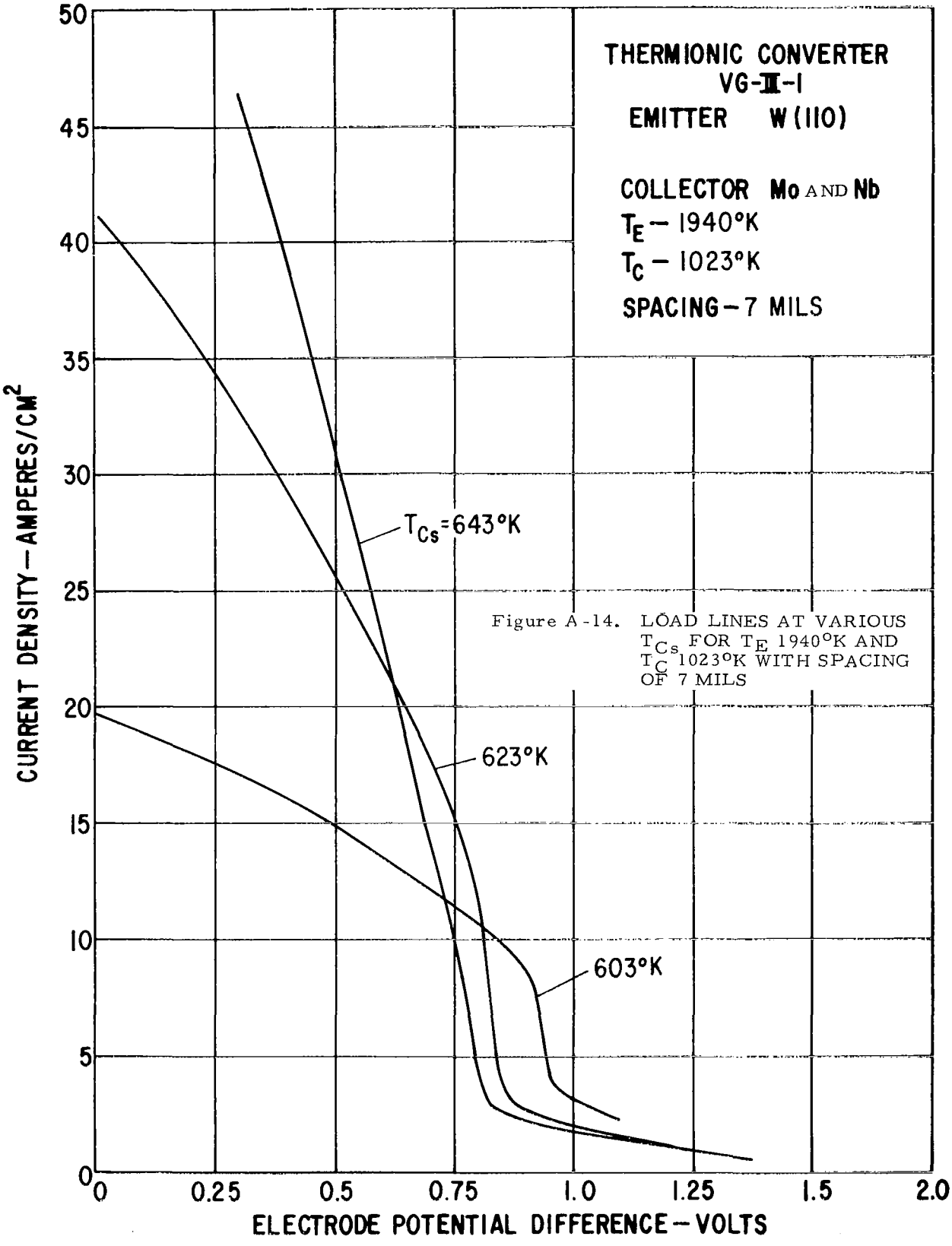

Figure A-14. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1940°K AND T_C 1023°K WITH SPACING OF 7 MILS

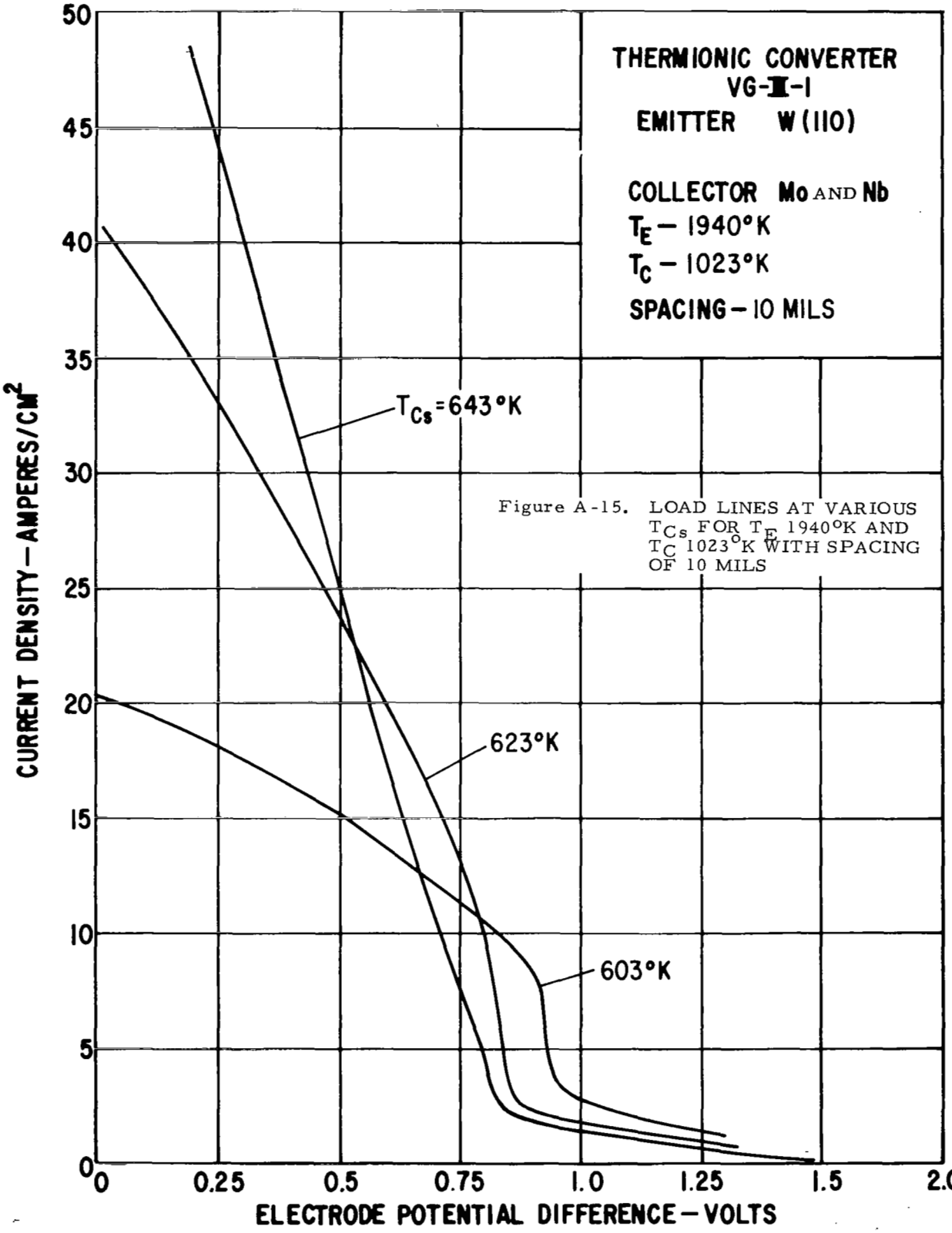

Figure A-15. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1940°K AND T_C 1023°K WITH SPACING OF 10 MILS

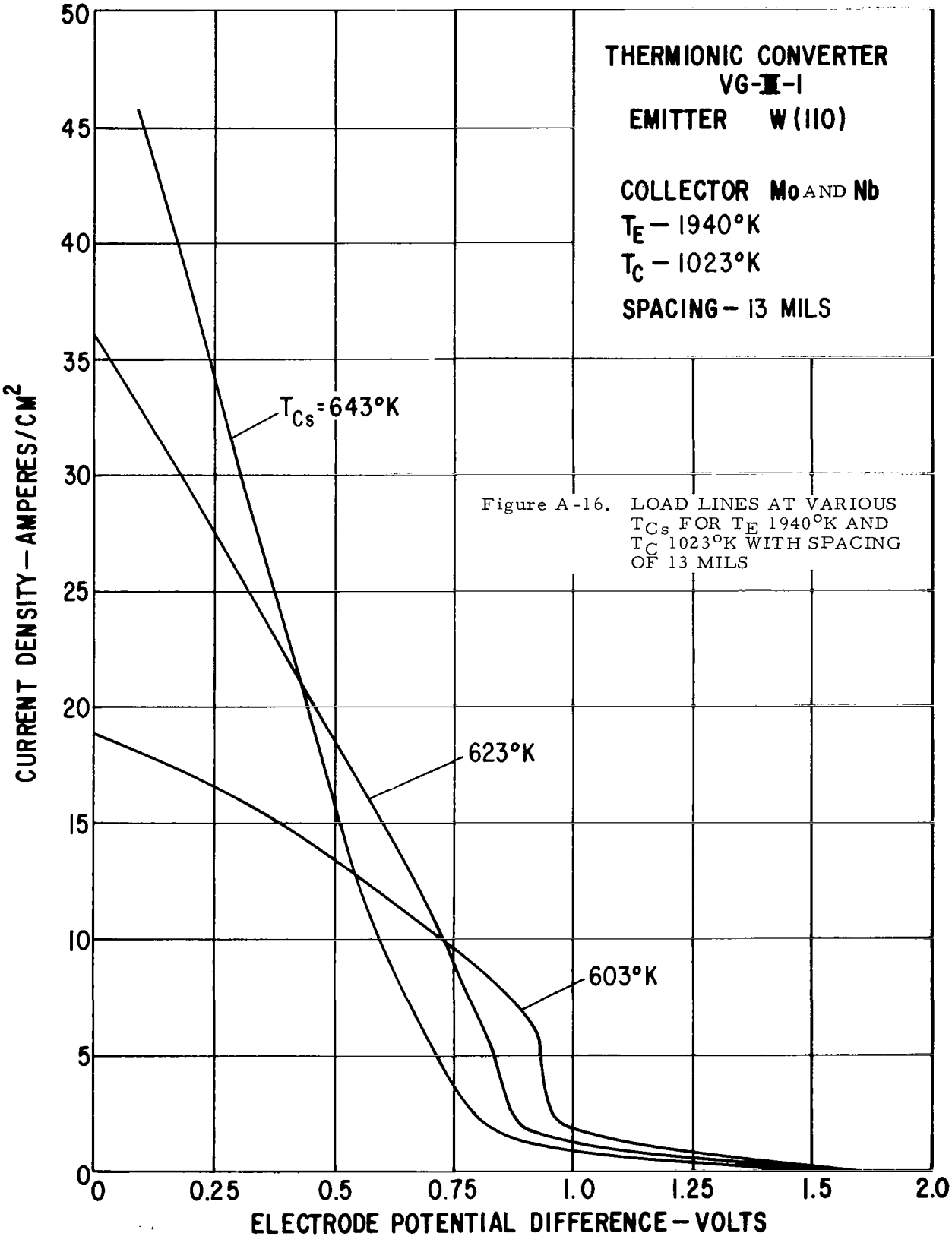

Figure A-16. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1940°K AND T_C 1023°K WITH SPACING OF 13 MILS

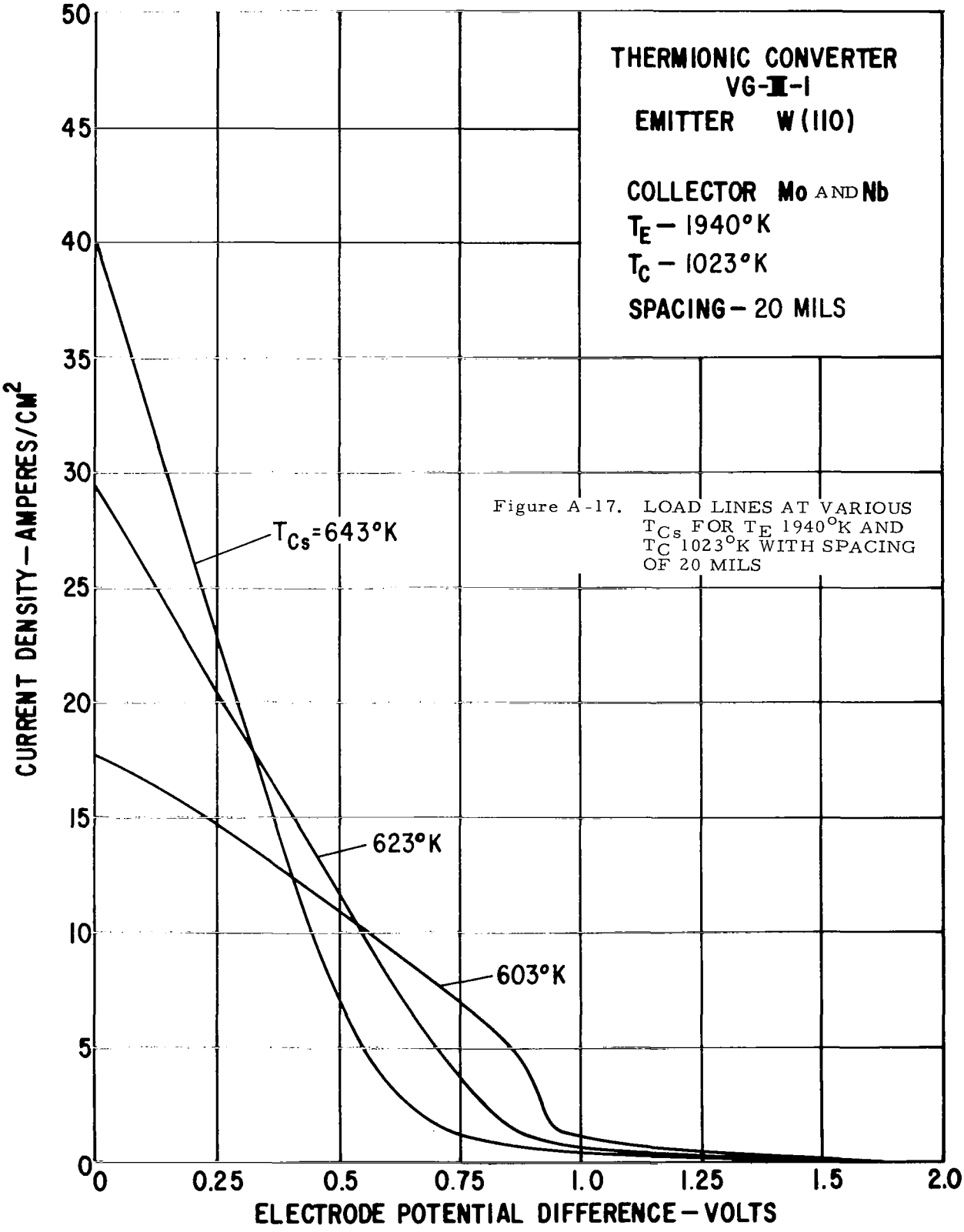

Figure A-17. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 1940°K AND T_C 1023°K WITH SPACING OF 20 MILS

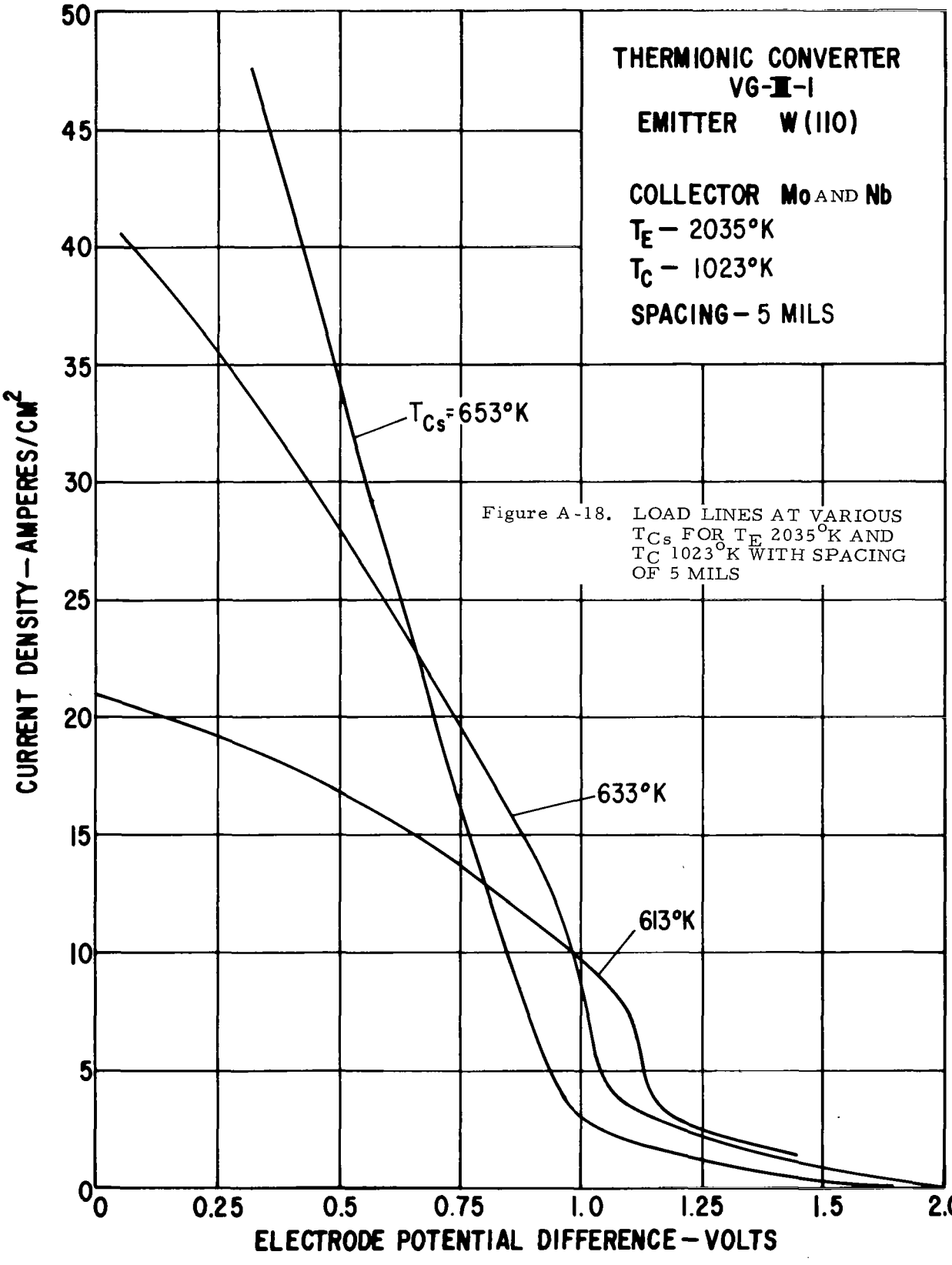

Figure A-18. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 2035°K AND T_C 1023°K WITH SPACING OF 5 MILS

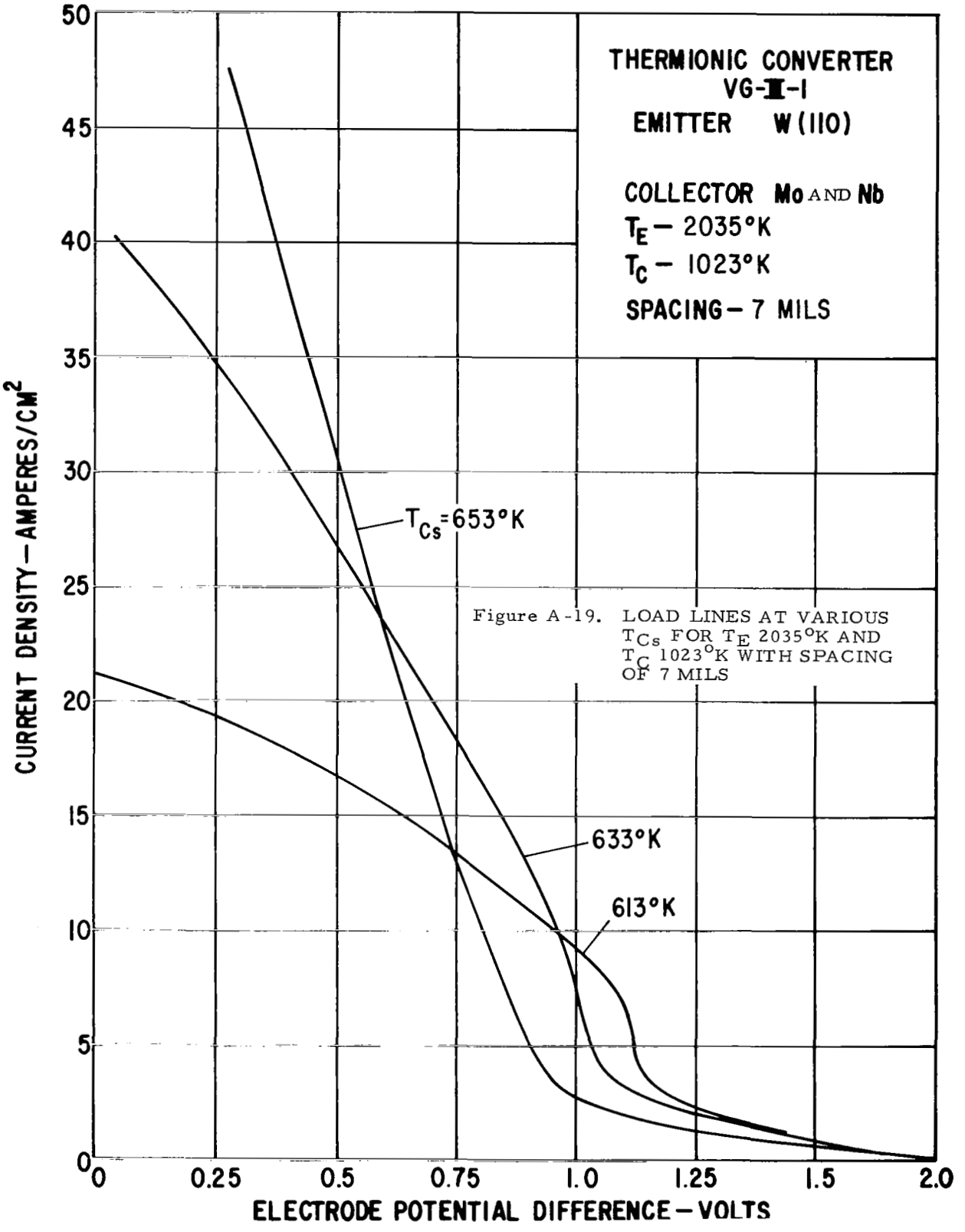

Figure A-19. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 2035°K AND T_C 1023°K WITH SPACING OF 7 MILS

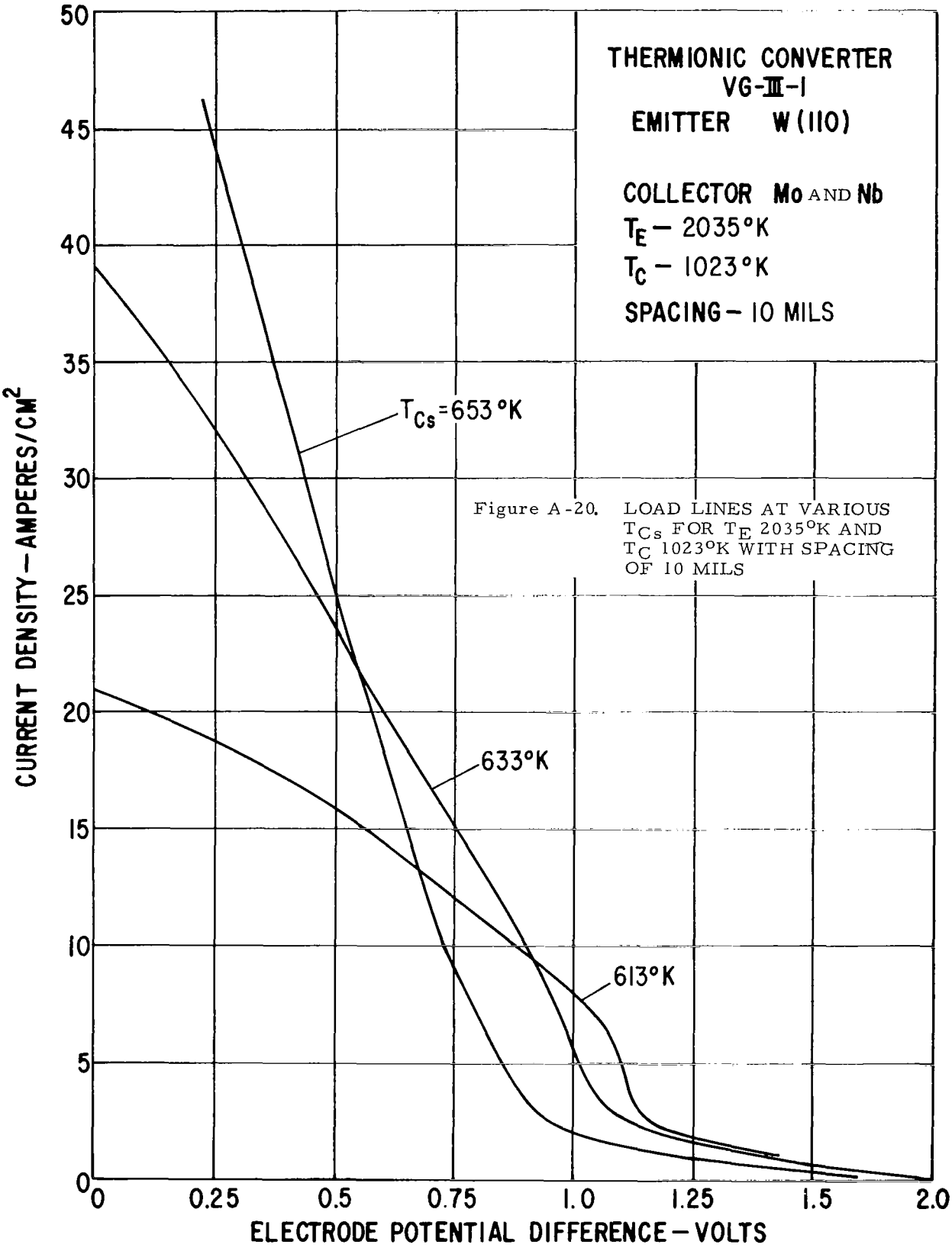

Figure A-20. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 2035°K AND T_C 1023°K WITH SPACING OF 10 MILS

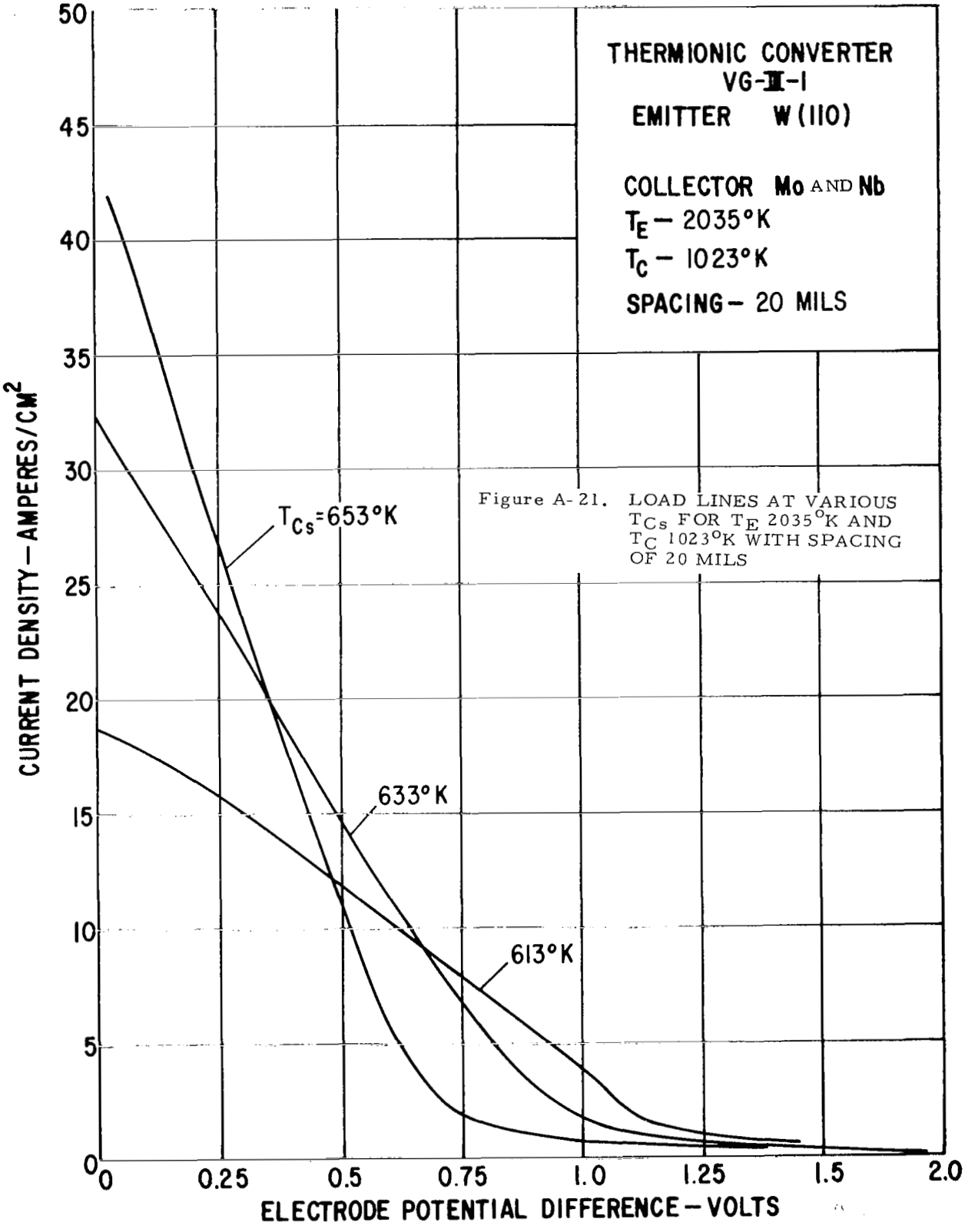

Figure A-21. LOAD LINES AT VARIOUS T_{Cs} FOR T_E 2035°K AND T_C 1023°K WITH SPACING OF 20 MILS

APPENDIX B

CALCULATIONS OF THERMAL GRADIENTS IN EXPERIMENTAL THERMIONIC CONVERTER EMITTERS

V. C. Wilson
General Electric Research and Development Center
Schenectady, New York

These calculations pertain to the emitters in the plane parallel thermionic converters listed in Table 1 that were built and tested at the General Electric Research and Development Center.

Figure B-1 is a cross section drawing of one of these emitters. It is 1 inch in diameter and 1/4-inch thick. Heat is put into the top surface by electron bombardment. Heat is removed from the bottom surface by radiation, Cs vapor conduction and electron and ion cooling. Also, heat is removed from the lower circumference by a thin refractory metal foil that serves as part of the cesium envelope, the emitter support and electrical lead. The temperature is measured by optical pyrometry of a black-body cavity that is shielded from receiving radiation from the hotter filament of the electron bombardment system. The path length of the heat flow is measured from the center of the black-body hole to the emitter surface.

The electron bombardment filament is wound with turns closer together at the outer edge to compensate for the heat removal by the supporting foil. Some measurements of the emitter temperature distribution have been published.[B1] Over the area of the collector, the emitter has a constant temperature within 5°C or less. With tungsten emitters, the temperature decrease to the edge is 25°C or less.

These calculations are for a correction to the emitter temperature due to the thermal gradient in the emitter. The

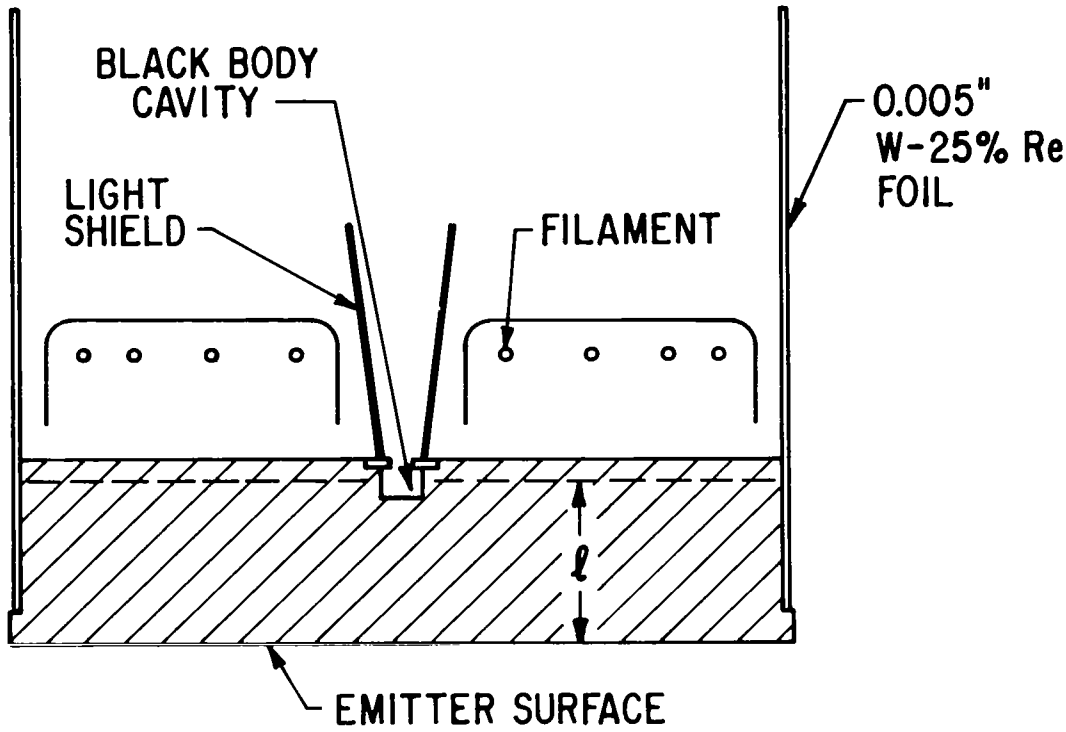

Figure B1. CROSS SECTION OF EMITTER, HEAT CHOKE, FILAMENT SHIELD AND LIGHT SHIELD

correction is less than 3%, and so a 20% error in the estimate would be an error of less than 0.6% in the emitter temperature. Therefore, no effort was made to estimate the radial flow of the heat to the edge of the disk. It was assumed that the heat flow from the depth of the center of the black-body cavity to the emitter surface was perpendicular to the emitter surface and of uniform density.

The calculations were based upon the sum of the heat removals from the emitter which were:

(1) Heat flow down supporting foil,
(2) Radiation,
(3) Electron and ion cooling (function of current density), and
(4) Cs vapor conduction.

(1) Heat Flow Down Supporting Foil

The heat that is removed by the supporting foil is radiated from a large metal structure. In an auxiliary test while outgassing one of the series (item 8, Table 1), the temperature of the radiator where it is joined to the foil was measured as a function of emitter temperature. From these data one can determine the ΔT in the foil at each emitter temperature. The heat flow was calculated from the simple equation $Q_L = A\ K\ \Delta T / \ell$ without corrections for radiation to or from the foil.

(2) Radiation from the Emitter Surface

For heat radiated between two parallel planes, an effective emissivity, $\epsilon' = (1/\epsilon_e + 1/\epsilon_a - 1)^{-1}$ may be used[B2] in the expression:

$$Q_r = \epsilon' \sigma (T_e^4 - T_c^4),$$

where ϵ_e is a function of T_E and ϵ_a is a function of $\sqrt{T_E T_C}$. Past experiences have indicated that well polished surfaces give an effective emissivity of ~0.16. Therefore, this value was used in these estimates.

(3) Electron and Ion Cooling

This term is a function of current density. A shunt was placed in the electrical lead to the emitter and the current determined by measuring the voltage across the shunt with a thermocouple voltmeter. This reads the effective heating of the current--i.e., the sum of the d.c. component and the root mean square of the a.c. component. Actually, one should have the d.c. plus the simple average of the a.c. component. The root mean square is 10% higher than the average. A Darsinval movement can be calibrated to read the average of a sign wave, but it ignores the d.c. component. Since in most of a thermionic converter testing program, the d.c. component is in the order of half the current, it was found best to use the thermocouple meter.

Dr. D. R. Wilkins of General Electric's Nuclear Thermionic Power Operation has developed a set of curves from computer calculations of the cooling by electrons and ions for various emitter temperatures, collector temperatures, current densities and Cs pressures.

Throughout a testing program for NASA, in order to compare each converter with those previously tested, emitter temperatures of 1665, 1770, 1865, 1962, 2057, 2153, and 2250°K were used. Wilkins made his calculations for emitter temperatures at each hundred degrees. Estimates of the electron cooling versus current density were made by interpolating between Wilkins' values. Calculations made using the data from Houston[B3] and Lawrence[B4] gave essentially the same results.

(4) Cesium Conduction

The heat removal from the emitter by the Cs vapor is independent of the current density and primarily a function of the emitter temperature and Cs pressure. Data from Kitrilakis and Meeker[B5] were used.

Having determined the sum of the cooling terms, the total heat flow per unit area through the emitter was used to calculate the thermal gradient in the emitter, ΔT. These values are shown in Figure B2, which plots ΔT versus metter readings for the emitter temperatures used. The cesium conduction correction is given in the table on the figure.

While operating a converter under test, the conditions for test, T_E, T_C, T_{Cs}, Load Resistance and a. c. drive were set. Then, the thermocouple meter was read. Using Figure B2, one could determine how much to increase the apparent black-body cavity temperature of the emitter to have the emitter surface at the desired temperature.

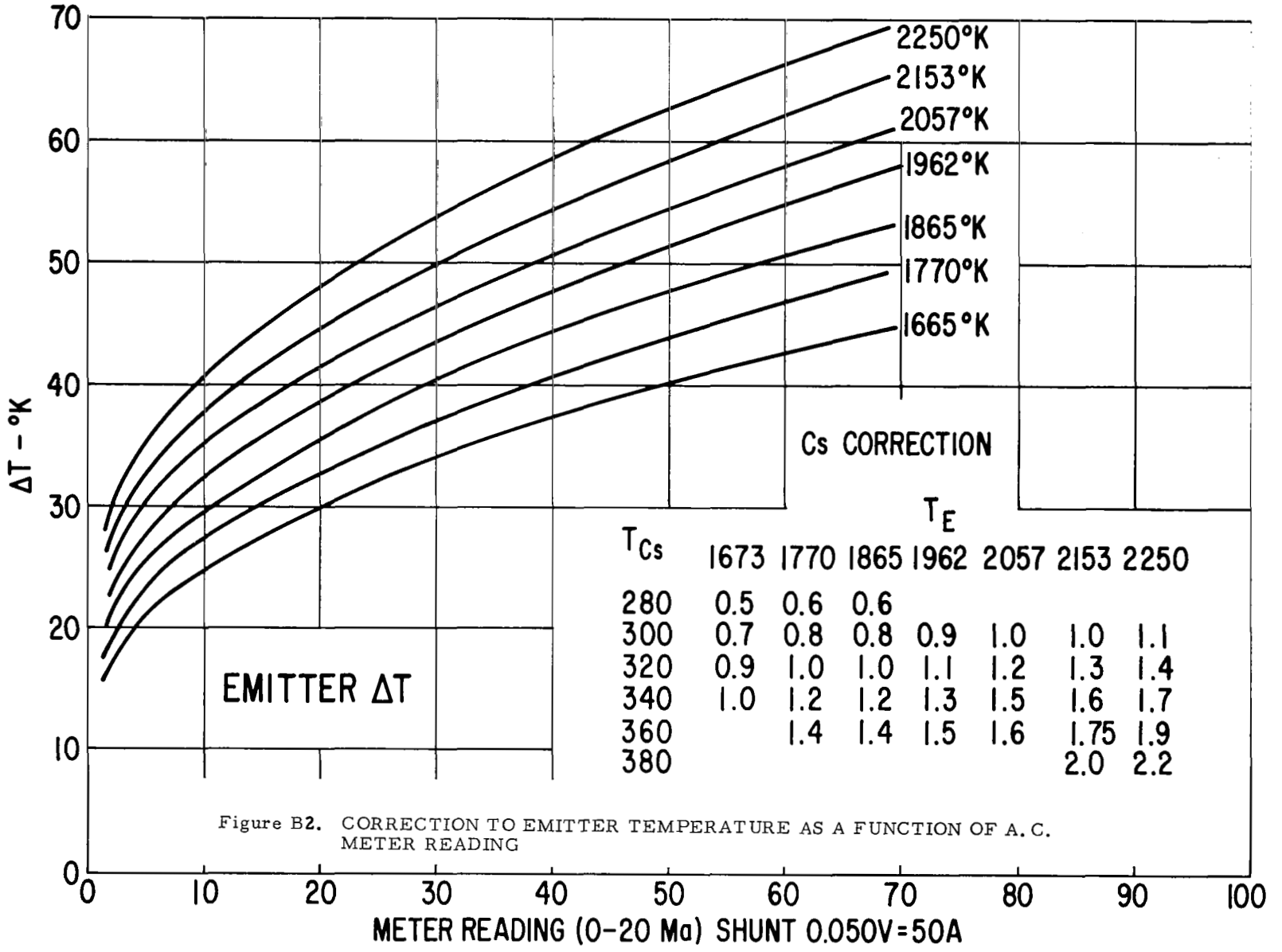

Figure B2. CORRECTION TO EMITTER TEMPERATURE AS A FUNCTION OF A.C. METER READING

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(B1) Wilson, V. C. and Lawrence, J., Adv. Energy Conv. 4, 201, Figure 4 (1964).

(B2) Block, F. G., Hatsopoulos, G. N., Wilson, V. C., Proc. Therm. Conv. Specialists Conf., San Diego, p. 379 (1965).

(B3) Houston, J. M., Therm. Conv. Specialists Conf., Gatlinburg, p. 214 (1963).

(B4) Lawrence, J., Therm. Conv. Specialists Conf., Cleveland, p. 291 (1964).

(B5) Kitrilakis, S. and Meeker, M., Adv. Energy Conv., 3, 59, (1963).

NASA-Langley, 1970 —— 22 E-5810