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Characteristics of a Plasma Thermocouple*

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The operation of a cesium plasma thermocouple is described for a range of hot-junction temperatures from 1600°K to 2600°K and for a range of cesium pressures from 10^{-5} mm Hg to 2 mm Hg. Electromotive force and short-circuit current data are presented for cells containing three different emitter substances: Ta, ZrC, and (ZrC)(UC). In the range of pressure and temperature variation studied, the observed electromotive forces are between 1 and 4.5 volts. Short-circuit current depends markedly on the current emission properties of the hot electrode. The largest short-circuit current density observed for the (ZrC)(UC) emitter, is 62 amperes per square centimeter.

INTRODUCTION

In a published letter,[1] the use of a plasma thermocouple as a practical heat-to-electric energy converter was proposed. The purpose of this article is to describe more fully some of the characteristics of this device with particular reference to the current density that can be achieved.

A generalized thermocouple can be considered to be made of a pair of joined, dissimilar, electrical conductors whose junctions are held at different temperatures. In the case of the plasma thermocouple, one conducting branch is a metal, the other is an electric plasma. Viewed in another way, the thermocouple is an elementary vacuum diode containing at low pressure some easily ionized gas. At the hot junction between metal and gas, the metal is a thermionic emitter. At the cold junction, the metal can be regarded as a current-collecting electrode. The alternative representations are shown in Fig. 1.

In a paper by Lewis and Reitz,[2] the operating properties of the cell are analyzed from a theoretical point of view. The cell electromotive force is described in terms of the characteristic thermoelectric power of the electric plasma. This is given, for the operating conditions of our cell, a value of about 1.3×10^{-3} volts per degree centigrade. Operated at a 2000°C temperature differential, an emf of 2.6 volts is expected, and the observed emf's are roughly in accord with this prediction. However, the detailed variation of electromotive force as a function of cesium pressure and of junction temperatures is certainly more complex than the prediction of this analysis. The observed electromotive forces for sample operating conditions are presented here.

Granting detailed variations in the electromotive force, which are not yet fully understood, an electromotive force between 2 and 3 volts is generally obtained, and this can probably not be increased in any substantial way by a variation of operating parameters. The sensitive measure of cell-power output is the current density which depends on the emitter temperature and, in some cases, the cesium pressure. The current characteristics of the cell are the chief subject of this article.

Lewis and Reitz identify four principal current limiting factors in the plasma thermocouple. The following summarizes their conclusions for reference in this paper.

(1) The current emission limit of the emitter electrode. When cell current densities approach the saturated emission of the hot electrode, then the emission characteristic becomes an important factor in current limitation. The saturated emission current density cannot be exceeded.

(2) Plasma resistance. The plasma offers an ohmic resistance to the cell current. The resistive mechanism is primarily multiple scattering of electrons by the plasma ions.

(3) Space charge. There must be a space charge region, where the plasma is not neutral, at each of the electrode boundaries.

(4) Cold layer. The cesium gas is well ionized (about 10% fractional ionization) throughout most of the

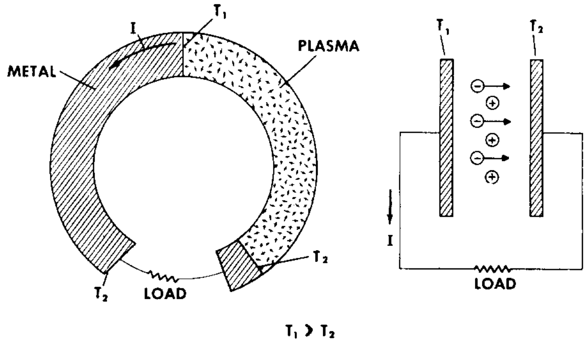

Fig. 1. Alternative representations of the plasma cell as a thermocouple and as a gas-filled diode.

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[1] Grover, Roehling, Salmi, and Pidd, J. Appl. Phys. 29, 1611 (1958).

[2] H. W. Lewis and J. R. Reitz, J. Appl. Phys. 30, 1439 (1959).

plasma body. However, at the cold junction there must be a gas layer which is poorly ionized. The lowering of ion density in this region is another factor in cell current limitation.

All four factors are discussed quantitatively by Lewis and Reitz.[2] Again, the observed cell currents are roughly in accord with their analysis. Experimental work directed toward an understanding of the cell operation in terms of the Lewis-Reitz analysis is made particularly difficult, however, by the fact that all four factors are in force, and it is difficult to separate their effects. In this article we single out as well as possible the first factor listed, the effect of emission limitation.

EXPERIMENTAL CELL

Figure 2 shows the arrangement of cell components. Two emitters are provided. Each is a tantalum disk of $\frac{5}{8}$-in. diameter and $\frac{1}{8}$-in. thick. The disks are welded onto 0.012-in. thick tantalum plates, and the plates form part of the vacuum-chamber wall. When different emitter substances, such as the carbides, are used, these are coated onto the $\frac{5}{8}$-in. tantalum disks, usually in a layer about $\frac{1}{32}$-in. thick. The tantalum plates are clamped in knife edges machined onto the flanges of the outer chamber. Such clamping pressure is applied so as to achieve a cold-weld joint. These joints provide excellent static-vacuum seals which can withstand the 300°C temperature excursions to which the whole chamber is subjected.

Halfway between the two emitter planes, and parallel to them, is a straight copper tube of $\frac{3}{16}$-in. outside diameter. Both tube ends are lead outside the chamber through porcelain insulator seals. Oil forced through the tube maintains it at nearly the same temperature as the chamber wall. The tube is the collector electrode of the thermocouple; the tantalum disks are the emitter electrodes.

During assembly, a sealed glass capsule containing liquid cesium metal is placed in the side tube. After the chamber has been evacuated, baked, and sealed, the capsule is broken by pinching the outer tube. The chamber is surrounded by a large bath of silicone oil which can be regulated in temperature from 40°C to 300°C, giving a cesium-vapor pressure range from 10^{-5} mm Hg to 2 mm Hg in the chamber.

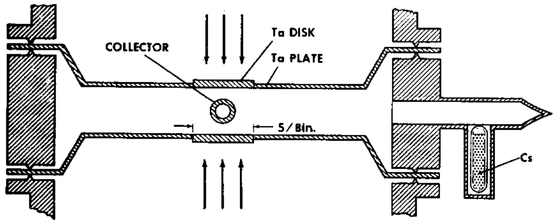

Fig. 2. Schematic drawing of the experimental cell chamber showing the emitter electrodes, collector electrode, and the cesium capsule inclusion prior to breakage.

Shown, only symbolically, are the electron beams which are used to bombard the tantalum disks from the outside. The bombardment capacity is 3 kv at 1 ampere. With the maximum power input, the tantalum disks can be raised in temperature up to 2900°C.

It should be pointed out that in the data quoted here only one emitter is used. Data have been obtained for two emitters acting simultaneously, but they are not sufficiently systematic to present at this time.

ELECTROMOTIVE FORCE

When a temperature difference is impressed between the two electrodes, a terminal potential difference appears. The data shown in Fig. 3 are obtained by placing a high-resistance voltmeter across the electrodes. All curves in Fig. 3 shown are for a tantalum emitting surface. The reading which is called the open-circuit voltage is a crude but useful measure of the cell electromotive force. Actually, true electromotive force is obtained by analyzing the complete voltage *versus* current relationships for the cell. However, these more exact data do not differ from the crude measurement by more than 10%. An article on voltage-current analysis is in preparation.

Curve (a) in Fig. 3 shows the open-circuit voltage *versus* emitter plate temperature for the case when cesium has not yet been admitted into the cell. The circuit resistance in this case, including leakage resistance in the insulators and the voltmeter resistance, is 2 megohm. The collector electrode is held at 40°C.

Curve (b) shows the open-circuit voltage when cesium is admitted to the cell at a chamber and collector temperature of 40°C. At this temperature the cesium pressure is 10^{-5} mm Hg, not much greater than the pressure of residual gas in the static vacuum. It can be seen, however, that the open-circuit voltage form is considerably modified by the addition of this small amount of cesium. Of course, the current characteristics are also drastically changed between cases (a) and (b). The maximum current for condition (a) is less than one milliampere, whereas for case (b), the current is one ampere or greater. A voltage maximum of about 2 volts in height occurs between the lowest point shown at 1400°C and 500°C. However, the plate temperatures are not observed in that range with sufficient accuracy to portray the temperature dependence.

Curve (c) shows the open-circuit voltage at a chamber temperature of 150°C corresponding to a cesium pressure of 10^{-2} mm Hg. The large inversion which can be seen is always characteristic of this cesium pressure value, and it has been observed in quite different cell configurations.

Curve (d) shows the open-circuit voltage at a chamber temperature of 250°C and a cesium pressure of 0.4 mm Hg. The inversion has nearly disappeared. This is the pressure range in which the cell is operated for best performance.

The analysis[2] of the electromotive force in terms of

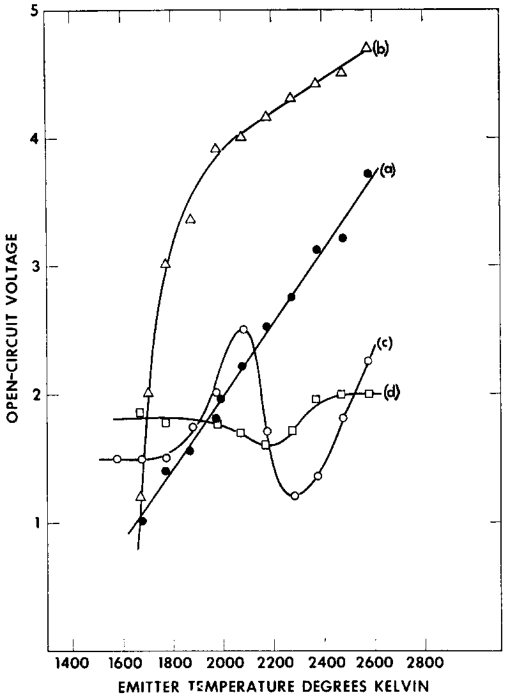

FIG. 3. Observed open-circuit voltage *versus* the tantalum emitter temperature in degrees Kelvin. Curve (a), vacuum cell with no cesium admitted; curve (b), cesium at a pressure of 10^{-5} mm Hg; curve (c), cesium at a pressure of 10^{-2} mm Hg; curve (d), cesium at a pressure of 4×10^{-1} mm Hg.

the plasma-thermoelectric power predicts a nearly linear relationship between voltage and temperature difference. Since large departures from linearity are observed for lower cesium pressures, it must be assumed that factors not contained in this analysis are important. We suspect that an important missing factor is represented by the electrical properties of the bounding electrodes. However, the analysis is not intended for use except at the highest cesium pressures, and at this point we no longer see the large voltage excursions.

CURRENT DENSITY

Short-circuit current, obtained by placing a low-impedance ammeter directly across the cell terminals, is a useful guide to cell performance. Roughly speaking, the current delivered to a matched external load is about one-half the short-circuit current. The basis for this kind of performance, and a description of deviations from the general rule, will be the subject of another paper now in preparation. Here we present only short-circuit current as an index of cell behavior.

In the series of Figs. 4 through 7, each figure shows a composite of two kinds of measurements. Before the plasma cell is assembled, the emitter plate and an associated electron-gun heater are placed in an auxiliary vacuum chamber for the purpose of measuring the saturated current emission as a function of emitter temperature. A disk collector of the same size as the emitter disk is placed 1 cm away from the emitting surface. A standard guard-ring configuration is added to provide a nearly uniform field between the emitter and collector planes. Accelerating potentials up to 3 kilovolts are applied to overcome space charge. The results of the vacuum emission measurements are shown as solid lines in the figures. An extension of the emission behavior beyond the highest point observed, about ½ ampere per square centimeter, is shown as a dashed curve. The extrapolation is made by using the two-parameter Richardson equation which best fits the data over the range of measurement. After the emission data are so recorded, the same emitter plate is assembled into a plasma cell, and short-circuit currents are observed by placing an ammeter across the cell terminals. The driving potential in this case is solely the thermal electromotive force of the cell. The short-circuit current data, normalized to current per square centimeter of emitter surface area, are shown as points on the graphs. The objective is to compare short-circuit current with saturated vacuum emission.

In Fig. 4 are shown a set of short-circuit current data for a cell which has a tantalum emitting surface. The short-circuit current data are recorded for three dif-

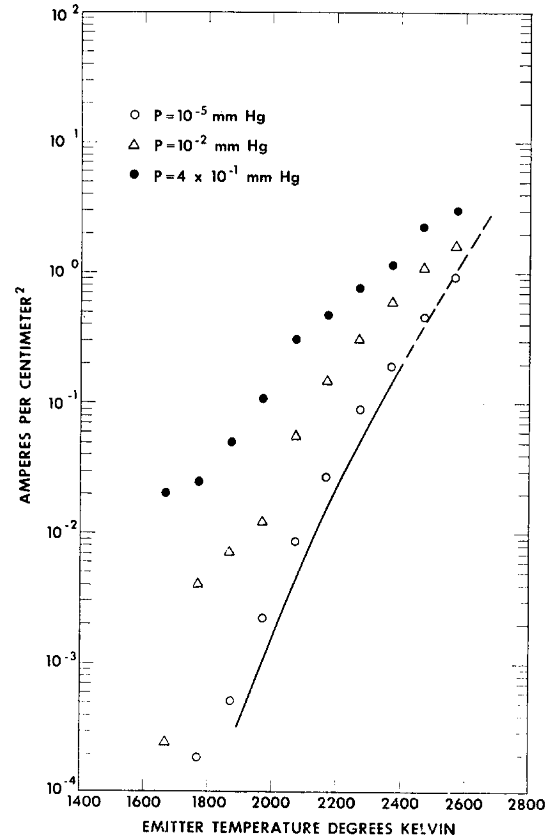

FIG. 4. Observed short-circuit current densities for a tantalum-emitter cell. The solid curve represents the measured saturated emitter current in vacuum. Dashed curve is an extrapolation of the vacuum emission data. Circle ○, cesium pressure is 10^{-5} mm Hg; triangle △, cesium pressure is 10^{-2} mm Hg; ●, cesium pressure is 4×10^{-1} mm Hg.

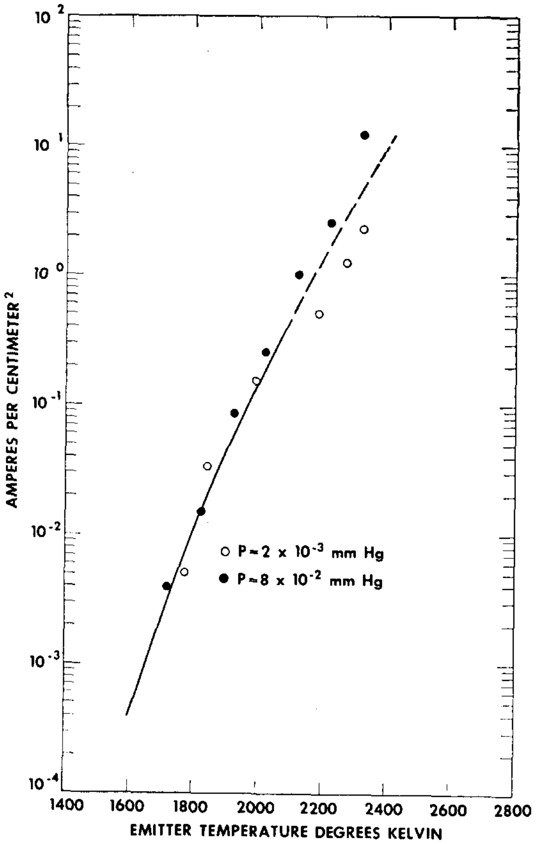

FIG. 5. Observed short-circuit current density for a cell with a zirconium-carbide emitter. Solid curve represents the measured saturated emission current in vacuum. Dashed curve is an extrapolation of the saturated vacuum emission data. ○, cesium pressure is 2×10^{-3} mm Hg; ●, cesium pressure is 8×10^{-2} mm Hg.

ferent cesium pressures in the cell. For the lowest pressure of 10^{-5} mm Hg, we find the short-circuit current dependence upon temperature to be almost identical with the saturated emission from the tantalum surface in vacuum. As the cesium pressure is increased, the cell currents exceed the saturated emission curve by a large factor. At a pressure of 4×10^{-1} mm Hg, short-circuit current exceeds saturated emission by a factor of 100 at 1800°K and by a factor 3 at 2600°K. It is evident that the presence of the cesium atmosphere enhances the current emission of tantalum, most likely in a mechanism similar to that described by Langmuir[3] for the emission of tungsten in the presence of cesium. In accordance with the Langmuir description, the current emission of the metal in the presence of cesium always approaches the vacuum dependence at high emitter temperatures.

It is difficult to draw conclusions about the relationship between short-circuit current and saturated emission in the case of the tantalum emitter. If the cell is emission limited, then the data shown should accurately portray the saturated emission of tantalum in the presence of cesium. However, any plasma resistance in the cell will suppress the current below the saturated level. Evidence is presented, however, supporting the point of view that the cell is fully emission limited at current densities below one ampere per square centimeter.

Both in an effort to improve cell performance and to gain a better understanding of the role of the cell emitter properties, two new emitter substances have been used in the plasma cell. These are zirconium carbide and a polycarbide, zirconium carbide: uranium carbide. The preparation, use, and characteristics of these substances as emitters are the subject of another paper.[4] A summary of their vacuum emission characteristics is to be found in the solid curves in Figs. 5–7. The primary advantages of the carbides for use in the cell are (a) high melting point, (b) stability in a cesium atmosphere, and (c) superior emission characteristics. The objective of the search for new emitters was to find superior intrinsic emission which is not limited by cesium pressure as in the case of the tantalum cell just described.

Figure 5 shows the short-circuit current for a cell with a zirconium-carbide emitter, with the saturated

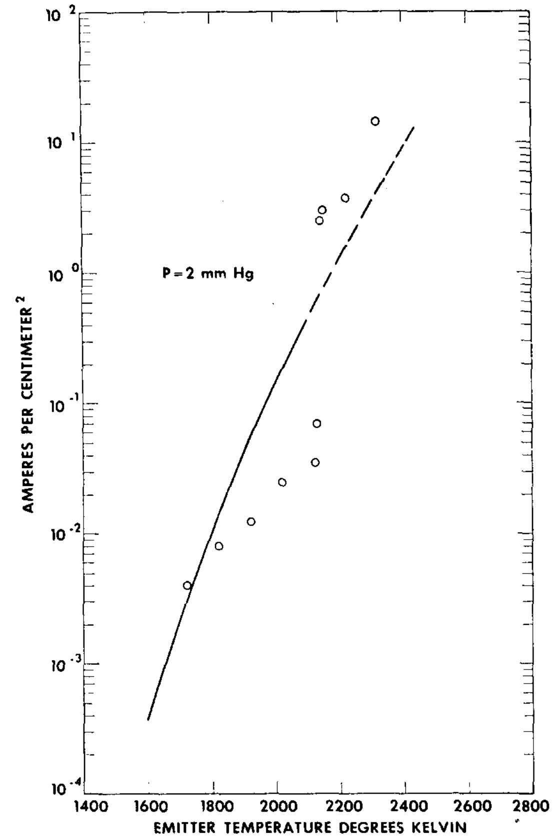

FIG. 6. Observed short-circuit current density for a cell with a zirconium-carbide emitter. Solid curve represents the measured saturated emission current in vacuum. Dashed curve is an extrapolation of the vacuum emission data. Circles represent the short-circuit current densities or a cesium pressure of 2 mm Hg.

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[3] I. Langmuir and K. H. Kingdom, Science 57, 58 (1923).

[4] Pidd, Grover, Roehling, Salmi, Farr, Krickorian, Witteman, J. Appl. Phys. 30, 1575 (1959).

vacuum emission shown for reference in the solid curve. For the two cesium pressures shown, it is evident that the short-circuit current under cell operation and the saturated vacuum current agree exactly. From this we draw two conclusions. One is that there is no marked Langmuir-type interaction between cesium and zirconium carbide which modifies the work function. Otherwise the current should be pressure dependent as in the case of the tantalum emitter. Two, the short-circuit current appears to be completely emission limited in the operating range portrayed.

Figure 6, also pertaining to a zirconium-carbide emitter cell, shows a more complex behavior. At the pressure 2 mm Hg the cell current at low temperatures is well below the emission curve. However, at 2120°K the current breaks upward, rising somewhat above the emission curve. The large current discontinuity is stable and accurately reproducible. No explanation is offered here for the sudden shift of current from one characteristic level to another.

The excellent emission characteristics of the polycarbide ZrC:UC are shown in Fig. 7. Trials of cell operation with this emitter are limited to the one pressure value, 3×10^{-1}, shown. Again, at current levels below 1 ampere per square centimeter, the short-circuit current data agree exactly with the saturated emission data. At 1600°K emitter temperature, the current is still no higher than that obtained with a tantalum emitter at $P=4\times10^{-1}$. However, at 2000°K, the ZrC:UC cell produces 40 times the current density of the tantalum cell operated under the same conditions. Thus, we conclude that the current in the tantalum cell at 2000°K is wholly limited by emission rather than by plasma resistance. Also, from the agreement between saturated emission and short-circuit current at values as high as 3 amperes per square centimeter, it is tempting to say that even in the ZrC:UC cell we are still primarily emission limited.

At 2100°K and above, the short-circuit current is substantially depressed below that expected from the saturated emission curve. This is the first direct demonstration of a plasma-limited current rather than an emission-limited current. It might be argued that the emission properties in this same range are not measured but extrapolated and that, therefore, the proof is not conclusive. However, the emission characteristic would have to be anomalous indeed to have the current-temperature relation change so drastically.

The highest short-circuit current observed at 2620°K is 124 amperes for the 2-cm² emitter area, or 62 amperes per cm². At this point the power density is about 30 watts per square centimeter, three times greater than the power density reported previously.[1] The estimated efficiency is also increased by a factor of three. It is now 15% as compared with the 5% figure previously reported.[1]

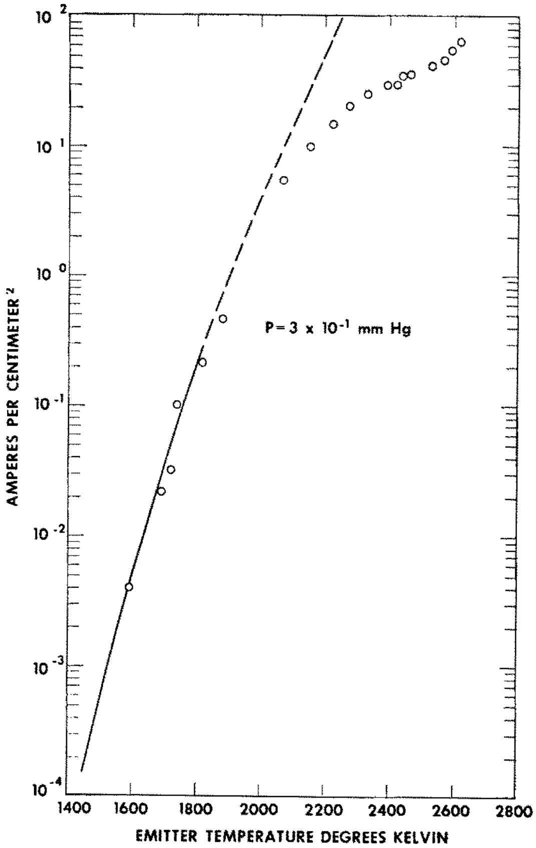

FIG. 7. Observed short-circuit current densities for a cell with a ZrC:UC emitter. Solid curve represents the measured saturated current emission in vacuum. Dashed curve is an extrapolation of the vacuum emission data. Data points represent the short-circuit current densities for a cesium pressure of 3×10^{-1} mm Hg.

CONCLUSIONS

In the introduction, four current limiting factors were listed, one relating to the emitter characteristic, and the rest pertaining to the plasma itself. Studies have been conducted on all these factors. The studies include variations of cesium pressure, variations of electrode spacing, and variations of collector size, material, and temperature. It is evident, however, in retrospect that emission limitation may have masked the results of these experiments. In the experimental program now in progress, the studies of the plasma-current limitation are being repeated in the higher current range, at 20 amperes per square centimeter and above, where it appears that emission is not the determining factor. Thus, the exploration of cell performance is not complete. The value of the work presented here is that of producing better guide lines for the research effort.

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