ANALYTICAL AND EXPERIMENTAL STUDIES OF THERMIONICALLY EMITTING ELECTRODES IN CONTACT WITH DENSE, SEEDED PLASMAS[†]

John K. Koester[*], Miklos Sajben[**], and Edward E. Zukoski[***]
Daniel and Florence Guggenheim Jet Propulsion Center
Kármán Laboratory of Fluid Mechanics and Jet Propulsion
California Institute of Technology, Pasadena, California 91109

Abstract

Interactions are considered between a moving, alkali-metal seeded, dense plasma and a metallic electrode whose surface properties are influenced by the absorption of seed particles. The plasma behavior is governed by a set of differential equations, which are coupled to the surface through the boundary conditions. These conditions are obtained by utilizing the particle desorption rate expressions of Levine and Gyftopoulos. The solution of the problem yields the state of the surface as well as the spatial distribution of plasma properties. In particular, electrode voltage drops are predicted, which indicate whether the electrode operates in a thermionic or arc mode. The method has been applied to a potassium-seeded argon plasma in contact with a tungsten electrode in a stagnation flow geometry. The results show that the plasma - surface interaction may lead to large electrode currents at moderate voltage drops. These currents can be up to an order of magnitude greater than what the random electron current would be at the surface under conditions of perfect thermodynamic equilibrium at the surface temperature. Results of a comparable experiment show reasonably good agreement with the theory.

I. Introduction

Metallic electrodes used in MHD devices conduct electricity by the streaming of internal free electrons. Currents in the plasma, on the other hand, have contributions due to the motion of both electrons and ions. By continuity, the total current cannot change across the plasma - electrode interface. On the cathode, this condition can be satisfied in two ways: (a) through the emission of electrons from the surface by some process (e.g. thermionic, field, or photoelectric emission), or (b) through the recombination of incident ions at the surface. Since in the bulk of the plasma electron currents are far greater than are ion currents, usually the emissive mechanism dominates. The recombination mechanism may become important if substantial ionization occurs in the cathode sheath, increasing the ion current at the expense of electron current.

In the case of thermionic emission, the emitted electron current is limited by the surface work function and surface temperature. Once the current passing through the electrode reaches this limit, no further increase of current can be accomplished by increasing the plasma - electrode voltage: any additional potential difference will simply increase the cathode voltage drop (V_c) that exists across a thin electric boundary layer adjacent to the surface, without appreciably influencing either the bulk of the plasma or the current passing through it. Experience also shows that at high values of cathode drop the electron emission will no longer be reasonably uniform over the surface; rather it will concentrate in small, intensely heated arc spots. The uniformly emitting cathode is commonly referred to as one operating in the thermionic mode, while the non-uniform emission is usually called an arc mode.

High cathode drops are undesirable in many MHD devices since they reduce the electrical efficiency and increase the heat loss to the wall. The presence of arc spots greatly accelerates the erosion and hence reduces the lifetime of electrodes, while further increasing heat losses due to the concentrated local heating of the walls. The large degrees of ionization in the vicinity of the spots consume useful energy and degrade performance accordingly. These factors all point to the need for electrodes capable of passing high currents at low cathode drops.

It is well known that alkali metals (commonly used to seed MHD generator plasmas) react chemically with the cathode surface to form an adsorbed layer. This layer reduces the effective work function of the surface, which in turn greatly increases the maximum electron emission rate for a given surface temperature.

It is the purpose of this paper to show that the complex interactions between a seeded, non-equilibrium plasma and an alkali metal-covered electrode are such that the thermionic mode of operation can be maintained at currents greatly in excess of the electron current emitted from the surface under conditions of thermal equilibrium. It is found that both the emissive and recombination mechanisms play important roles in bringing about this situation.

The theory presented in this paper is a result of a merger between results taken from sur-

[*] Assistant Professor of Aerospace Engineering, University of Tennessee, Space Institute

[**] Assistant Professor of Aeronautics, California Institute of Technology

[***] Professor of Engineering and Jet Propulsion, California Institute of Technology

[†] Supported in part by the U.S. Air Force Office of Scientific Research under Contract AF 49(638)-1285.

face and plasma physics, which allows a prediction of cathode voltage drops as a function of surface temperature and total electrode current for a specified plasma composition, temperature, and density. By requiring the cathode drop to be sufficiently low, quantitative conditions are delineated within which a cathode is expected to operate in the thermionic mode.

The theory assumes the plasma to be sufficiently weakly ionized to make the heavy particle flow and temperature fields uncoupled from the motion of seed particles, so that they may be initially specified. Finite rate volume ionization and recombination is taken into account.

The theory has been applied to a tungsten cathode in a stagnation flow geometry, a situation which could be conveniently duplicated experimentally. The experimental results agree with the predictions of the theory within a factor of two for conditions where a low cathode drop ($V_c < 10$ V) is predicted. If V_c is predicted to be high, then certain assumptions of the present theory are violated and the theory is expected to break down. The experiments indicate that under such conditions the cathode works in the arc mode. The existence of concentrated emission is not predicted by this theory, but its breakdown is a convenient indication of the possible development of arc spots.

II. General Description of Coupling Effects

It is known [1] that nearly all seed particles, whether charged or not, are adsorbed to the surface, due to the large affinity between alkali metal and the metallic electrode. This adsorption results in partial or complete coverage of the surface, even if the partial pressure of the seed is below the saturation pressure determined by the wall temperature. The surface layer, characterized by the degree of coverage (θ), determines a greatly modified work function. Together with the wall temperature (T_w), θ also determines the rate of emission for electrons (ν_e, no. /cm^2, sec) and the rates of desorption for ions and atoms (ν_i and ν_a, respectively). These emission rates can be calculated for any refractory metal partially covered with monatomic metallic particles as functions of θ and T_w, using the relations given by Levine and Gyftopoulos[2-5]. Application of these relations to the case of potassium-covered tungsten yielded the electron emission S curves of Fig. 1, which also shows all available experimental data. Note that the parameter is atom arrival rate, as customary, and that the high current data were obtained in the present experiment. The comparison is favorable over nine orders of magnitude of current densities, although the low current density part of the curves is displaced by about 100°K. The good agreement increases our confidence that the ion and atom desorption rates (for which no comparable experimental results seem to be available) will also be correct.

If the plasma is thought to be in thermodynamic equilibrium with the surface at the temperature T_w (i.e., the net flux of any species to the wall is zero), then the degree of coverage is determined solely by balancing the incident and desorbed seed atom fluxes at the surface[9]: $\nu_a(\theta, T_w) = \frac{1}{4}\epsilon_a n_c(T_w)\overline{C}_s(T_w)$; ϵ_a being the ratio of seed atom to carrier atom densities, n_c the number density of carrier atoms, and $\overline{C}_s$ the mean thermal speed of seed atoms. Having determined θ from this implicit, transcendental relation, the emitted electron current density can be computed as $J_{ez}(T_w, \epsilon_a) = e\nu_e(T_w, \theta)$. A $J_{ez}(T_w, \epsilon_a)$ plot, such as the one shown on Fig. 2, may be looked upon as the high pressure equivalent of electron emission S curves familiar from low-pressure plasma work. The main difference between the two is that in the latter case the arrival rate is considered an independent parameter, whereas on Fig. 2 the arrival rate is dependent on T_w, and ϵ_a is considered to be independent. Figure 1 was drawn for atmospheric plasma pressure. At low temperatures, the surface is almost fully covered and the work function is that of pure potassium (denoted as "K limit"), whereas at high temperatures the surface is practically bare and the work function of tungsten is the controlling parameter ("W-limit"). In between, the competiton between rising temperature and decreasing coverage results in a current maximum and minimum as shown.

J_{ez} is a convenient reference value for emitted currents. It should be remembered that it corresponds to equilibrium coupling between plasma and surface, which implies zero net current; the emission is exactly balanced by the electron flux arriving from the equilibrium plasma.

If the temperature of the bulk plasma deviates from the wall temperature, the arriving random atom flux still depends on T_w, since in the dense plasmas considered here the thermal boundary layers are much thicker than the mean free path, and therefore the gas and wall temperatures at the wall are the same.

If a voltage is imposed externally between the plasma and the electrode, the arrival rate of the (uncharged) seed atoms is not directly influenced, so one might expect that the coverage, and hence the surface work function, also remains unchanged. If it would, then J_{ez} would represent the maximum possible electron current at the given T_w temperature: the electrode characteristic curve would saturate and the cathode drop V_c would approach infinity at this current. However, this is not what actually happens.

In equilibrium, the incident and emitted fluxes of each individual species are strictly balanced. If a net current flows, one can only say that in steady state there can be no net accumulation of seed particles at the wall, and therefore the net flux of heavy particles to the wall must balance the net flux of heavies away from it. This condition quickly leads to the requirement

$$\Gamma_{aw} = -\Gamma_{iw} , \tag{1}$$

where Γ_{sw} is the net flux of the s species at the wall (s = a, i, e for seed atoms, seed ions, and electrons). This means that there is, in general, a net seed atom flux, and hence a neutral seed atom density gradient near the wall and that the diffusion of atoms is an integral part of the entire problem. The atom diffusion equation must be solved simultaneously with the plasma - surface problem, subject to the appropriate boundary conditions, eq. (1) being one of these.

It follows that in the case of a net current, the degree of coverage on the cathode surface is incluenced not only by the atom flux but also by the collected ion flux. Although the ion flux may be much smaller than the electron flux at the cathode, the coverage can nevertheless be increased considerably, since the incoming ion flux is counterbalanced only by the outward diffusion of seed atoms, which may be quite slow. Furthermore, the ion flux at the surface can be increased greatly over its value in the bulk plasma due to convection and ionization effects in the ambipolar (charge-neutral) region of the electric boundary layer.

A second, minor factor contributing to the increase of maximum possible current in the case of nonzero net currents is the Schottky effect. The large positive space charge in the sheath (charge-separated) region of the electric boundary layer gives rise to large electric fields at the wall (E_w). These fields may be large enough to increase the emitted electron flux due to the Schottky effect. In this case, ν_e is given as $\nu_e(\theta, T_w)A_E(E_w, T_w)$, where A_E is the Schottky correction.

In order to give a preliminary idea about the tremendous influence of these effects, Fig. 3 displays pictorially the cathode voltage drop V_c as a function of J_c (the net electron current) and T_w. Figure 3 is an end result of the present theory, taking into account all the above discussed effects, which clearly represent non-equilibrium coupling.

If the coverage would stay at the equilibrium value, then V_c would go to infinity at $J_c(T_w) = J_{ez}(T_w)$, in accordance with our arguments presented before. In graphic terms, this means that a vertical "wall" would appear on Fig. 3, built onto the $\epsilon_a = .004$ curve of Fig. 2. One could not increase J_c over this value, no matter how large voltage would be imposed across the plasma - surface system.

The combined action of increased coverage due to incident ions and Schottky effect changes this situation drastically. At about 1600°K, a gap is cut in the "wall," and over about 10 amps/cm^2 an entire new region of low cathode-drop (i.e., thermionic) operation comes into existence. This region would be wholly absent without non-equilibrium coupling effects. It has a far-reaching significance from the viewpoint of practical MHD generators: it means that (a) efficient thermionic operation is possible at large currents, and that (b) by the simultaneous control of wall temperature and total current, this large current regime can be reached after start-up without even temporary operation in the arc mode.

The $V_c(J_c, T_w)$ function depends sensitively on the choice of collision cross-sections required to calculate relevant diffusion coefficients. For some choices the gap may not exist, but a high current thermionic region always seems to be present.

III. Description of Analytical Work

It will be assumed that all mean free paths are much smaller than the extent of the charge-separated (sheath) region. Furthermore, the sheath is assumed to be thin compared to the electrical boundary layer, within which appreciable variation of charged particle density occurs. It is noted that the sheath thickness can be several orders greater than the Debye length based on bulk plasma properties ($h_{e\infty}$). A sheath thickness is, however, correctly estimated by the Debye length, if it is based on some representative local electron density within the sheath. Failure to distinguish between these two scales led to erroneous justification of the "free-fall" sheath models by some authors in the past.

The smallness of the mean free path justifies the continuum formalism. The assumptions of steady state, incompressible fluid, no magnetic fields, lead to the following equations governing the plasma phase, in MKS units:

Conservation of Species

$$\vec{u}\cdot\nabla n_s + \nabla\cdot\vec{\Gamma}_s = \dot{n}_s \qquad (s = i, e, a) \tag{2}$$

where $\vec{u}$ is the known viscous, neutral flow field, $\vec{\Gamma}_s$ is the flux of the s species, and $\dot{n}_s$ is the net production rate of s species per unit volume.

Particle Flux Equation

Conservation of momentum is accounted for by the particle flux equation:

$$\vec{\Gamma}_s = -D_s\left(\nabla n_s + \frac{q_s}{kT_s} n_s \nabla\phi\right) \tag{3}$$

where $q_i = e$, $q_e = -e$, $q_a = 0$, and D_s is the mutual diffusion coefficient of the s species. The mobilities have been related to the diffusion coefficients by Einstein's relation. It should be noted that eq. (3) is valid only if the species distribution functions deviate from Maxwellian only mildly, which will not be true for electrons and ions in a very strong electric field.

Poisson's Equation

$$\epsilon_o\nabla^2\phi = -e(n_i - n_e) \tag{4}$$

where ε_o is the permittivity of free space, ϕ is the electric potential, and only singly ionized positive ions are considered.

The ions and neutrals are assumed to be in translation equilibrium at the neutral temperature, T_n, while the electrons and excited states are in thermal equilibrium at a different temperature, T_e. T_e is governed by the electron energy equation, and its value in the bulk plasma was determined following the procedures used by Cool[6]. Due to the large electron thermal conductivity, the electron temperature stays constant at the bulk value throughout the electric boundary layer for sheath voltage drops less than about two volts. The electron energy equation in the electric boundary layer will therefore be conveniently replaced by the $T_e = T_{e\infty}$ = constant assumption. The electron production term for three-body recombination and thermal electron impact ionization is

$$\dot{n}_e = \gamma(T_e)n_e[n^2_{Saha}(T_e)-n_e n_i],$$

where n_{Saha} is the electron number density evaluated by the Saha equilibrium relation. The recombination coefficient, γ, is given by Cool[6] for potassium. The ion and atom production terms are directly related by $\dot{n}_e = \dot{n}_i = -\dot{n}_a$.

Using these production terms, eqs. (2), (3), and (4) can be reduced to four coupled partial differential equations for the potential and the three species densities. This system of equations must be complemented by an appropriate set of boundary conditions. For simplicity, we shall keep a flat plate or a stagnation point boundary layer situation in mind.

The conditions at the outer edge of the electric boundary layer are those corresponding to the bulk plasma. Using Cool's calculations, values of $n_{e\infty} = n_{i\infty}$, $n_{a\infty}$ and E_∞ were determined for each value of current density and used as outer boundary conditions.

The boundary conditions at the wall represent the surface - plasma interaction, and have been extensively discussed in ref. 9. In essence, these boundary conditions are arrived at by a reasoning as follows.

The emission rate of s species from the surface is determined by the surface temperature and the surface coverage in a manner known from the work of Levine and Gyftopoulos, i. e., $\nu_s(\theta, T_w)$ is given. The incident flux (μ_s) is postulated as a displaced Maxwellian[8], characterized by the surface density and net flux as parameters. The net flux away from the wall is thus given in a microscopic description as $\Gamma_{sw} = \nu_s\mu_s$.

More than several mean free paths away from the wall the continuum description must hold. Equating the continuum expression of flux to the microscopic expression obtained previously, algebraic relations are obtained in the form

$$\nu_s(\theta, T_w, E_w) = f(n_{sw}, T_w, \Gamma_{sw}), \quad s = e, i, a \qquad (5)$$

which must be satisfied at the surface and therefore represent the desired boundary conditions. Specification of the potential at the wall raises to eight the total number of boundary conditions available. This would be appropriate to the original problem (which is second order in each variable), were it not for the fact that eq. (5) introduces θ as an additional unknown. However, the additional condition necessary has already been given by eq. (1) discussed earlier, and the set of boundary conditions is thus complete.

The solution of the problem is far too complex to be even outlined in this paper. It involves a generalization of the asymptotic techniques developed for continuum probe theories by Lam[7], Bienkowski[8], and others, as well as several nested numerical iterations by computer. The interested reader is referred to Koester's work[10] for details.

IV. Analytical and Experimental Results for Stagnation Flow

The stagnation flow configuration was found to be convenient from both experimental and theoretical points of view. Laminar stagnation flow implies an electric boundary layer of constant thickness, which greatly simplifies both the analytical and computational work. The theory of Section III was applied to this case and the results were cast into the form of V_t versus J_t characteristics, where V_t is the total electrode voltage drop and J_t is the total current through the electrode. V_t is the sum of V_{sh}, the sheath voltage drag and V_{am}, the drop across the ambipolar portion of the electric boundary layer. The gas temperature was taken to be 2,000°K and the free stream velocity 100 m/sec. A range of surface temperatures and seed fractions were considered, corresponding to experimental limitations.

A typical computed current - voltage characteristic for T_w = 1,500°K and ε_a = .004 is shown on Fig. 4. The ambipolar voltage drop stays nearly constant at approximately one volt for all values of current, while the sheath drop is quite small everywhere except in cathode operation for currents greater than 8 amps/cm^2. Near this saturation current, the sheath drop increases rapidly to high values and accounts for most of the total drop V_t. At still higher currents, V_{sh} and V_t again decrease and level off at approximately ten volts. This is the same behavior as the one shown on Fig. 3: the cathode portion of Fig. 4 closely corresponds to an intersection of the T_w = 1,500°K plane with the three-dimensional surface of Fig. 3. The level portion of Fig. 4 over 8 amp/cm^2 cathode current represents the large current thermionic operation made possible by the non-equilibrium plasma-surface coupling.

The experimental apparatus shown on Fig. 5 was designed to meet the conditions of the theory as closely as possible. A potassium-seeded argon plasma, produced by an arc jet heater apparatus (described in ref. 6) flowed through the boron nitride tubular test section and exhausted at atmospheric pressure. The electrode voltage drop was measured by extrapolating the voltages measured by floating voltage probes to the tungsten electrode surface. The voltage probes were corrected for floating voltage drop by an approximate analysis which led to corrections up to one volt. The electrode surface temperature was measured optically through the quartz window.

Cathode temperature was not strictly controlled in these experiments: it was allowed to vary at some rate determined by the balance between the arc heating and the cooling provided at the back side of the electrode by an argon coolant stream. The coolant flow rate could be adjusted to serve as a rough control for T_w. This arrangement yielded both J_c and T_w as functions of time. A point representative of the state of the cathode on a J_c versus T_w plane thereby moved along some trajectory for any given run in which J_c was varied. Since the variation was very slow compared to times within which θ could change, the process was quasi-steady and the shape of the trajectories is therefore of no importance.

An experimental current - voltage characteristic, obtained from this device, is compared with theory in Fig. 6. The behavior for low values of current density compares favorably with theory, since the voltage drop discrepancy is of the same order as the approximate floating voltage probe correction. The experimental characteristic has a shallower shape than predicted as saturation is approached. This saturation delay indicates that some important secondary effect exists, probably electron heating in the electric boundary layer. Nevertheless, the saturation current densities are predicted within a factor of two. At cathode drops of around 10 volts, the electric boundary layer exhibits an instability which results in the formation of a readily observable tiny arc ("arclet") on the cathode surface. These arclets were bright spots near the surface, but became diffuse away from the wall, so that the bulk of the plasma remained reasonably uniform even after the formation of arclets.

The present theory does not allow for such three-dimensional solutions, and hence the arclet breakdown voltage is not predicted. The current at which such a breakdown occurs, however, is quite close to the saturation current predicted.

The regions of current density and cathode surface temperature for which cathodes may be operated in the low voltage thermionic mode may be determined from Fig. 7 for $\epsilon_a = .004$. The dotted lines indicate experimental trajectories; the wavy lines indicate that arclets were observed. The trends indicate that the thermionic mode for $T_w > 1,500^{\circ}K$ and $J_c > 10$ amperes/cm^2 is limited only by complete seed ionization or the thermionic emission from a pure potassium surface. As displayed on Fig. 7, the predicted high values of current density in the thermionic mode are verified by the experimental results. The low values of cathode drop around $T_w = 1,600^{\circ}K$ and $J_c =$ 9 amps/cm^2 were also observed experimentally. The 10-volt data points are in good agreement with the 10-volt level contours for cathode drop. Note the arclet behavior observed in the regions of predicted large voltage drops.

Similar behavior is found at lower seed fraction values (see Fig. 8 for $\epsilon_a = .002$). However, the cathode drop values in the "gap" or "channel" region are greater. The channel region is quite narrow in this case and sensitive to small changes in seed fraction. This sensitivity explains the increase in experimental scatter for the data in this case.

V. Conclusions

A method has been developed to describe the close interaction between a seeded plasma and a seed-covered metallic electrode. The theoretical predictions were found to agree with experiment within the expected margins of uncertainty. Both theory and experiment demonstrate the feasibility of operating the electrode at large currents while uniform emitted current distributions and low cathode drops are maintained. The theory also indicates how arcing might be avoided, both in steady state and in starting transient operation.

References

1. Langmuir, I. and Taylor, J. B., Phys. Rev., 44 (1933), pp. 423-458.

2. Gyftopoulos, E. P. and Levine, J. D., J. Appl. Phys., 33 (1962), pp. 67-73.

3. Levine, J. D. and Gyftopoulos, E. P., Surface Science, 1 (1964), pp. 171-193.

4. Levine, J. D. and Gyftopoulos, E. P., Surface Science, 1 (1964), pp. 225-241.

5. Levine, J. D. and Gyftopoulos, E. P., Surface Science, 1 (1964), pp. 349-360.

6. Cool, T. A. and Zukoski, E. E., Phys. of Fluids, 9 (1966), pp. 780-796.

7. Lam, S. H., AIAA Journal, 2 (1964), pp. 256-263.

8. Bienkowski, G. and Kalnavarns, J., AIAA Paper 68-167 (1968).

9. Sajben, M., "Boundary Conditions for Adsorbing - Emitting Electrodes in Contact

with Seeded, Dense Plasmas," accepted for publication in the AIAA Journal (1970).

10. Koester, J. K., "Analytical and Experimental Studies of Thermionically Emitting Electrodes in Contact with Dense, Seeded Plasmas," Ph. D. Thesis, California Institute of Technology (1970).

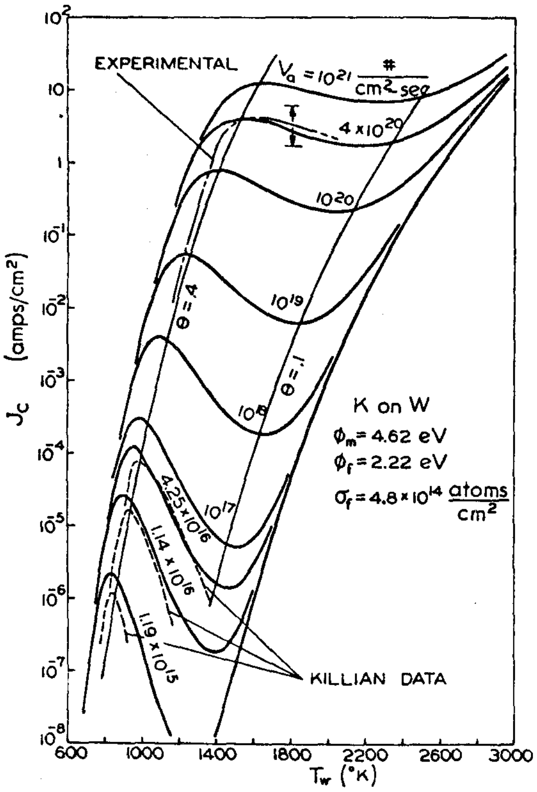

Figure 1. The Electron Emission S-curves Computed for Potassium by the Levine and Gyftopoulos Model, Compared with Available Experimental Data.

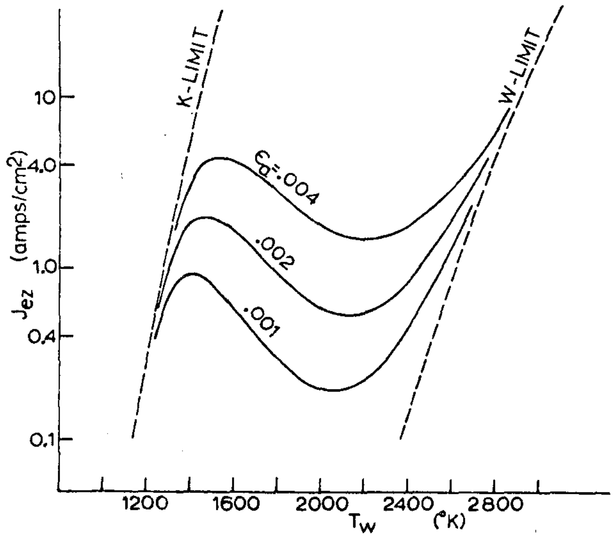

Figure 2. Thermionic Current Density Versus Surface Temperature for an Atmospheric, Potassium-seeded Argon Plasma on a Tungsten Cathode.

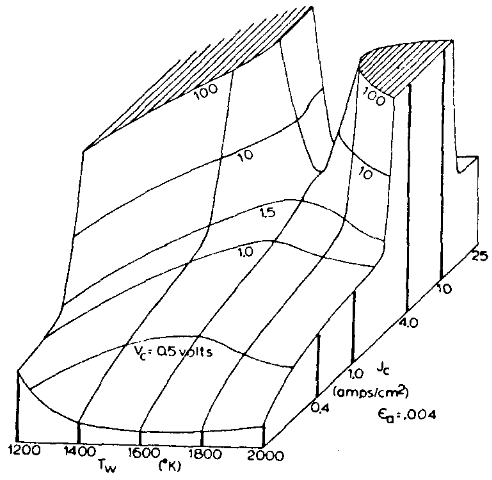

Figure 3. Cathode Voltage Drop Versus Surface Temperature and Current Density for the Potassium-Argon-Tungsten System.

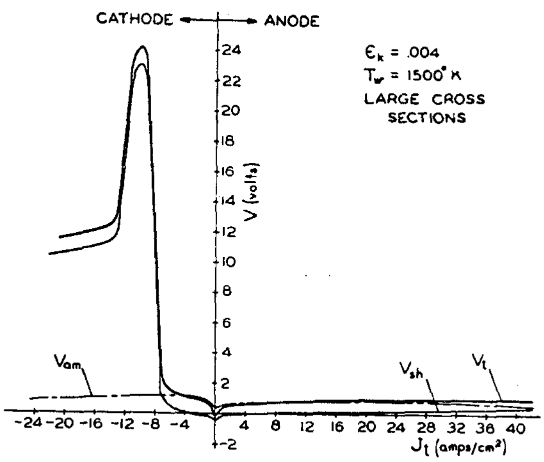

Figure 4. Current-Voltage Characteristics for the Stagnation Flow Electrode.

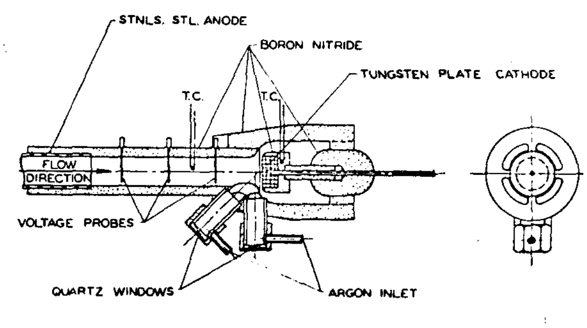

Figure 5. Tubular Test Section with Stagnation Flow Cathode.

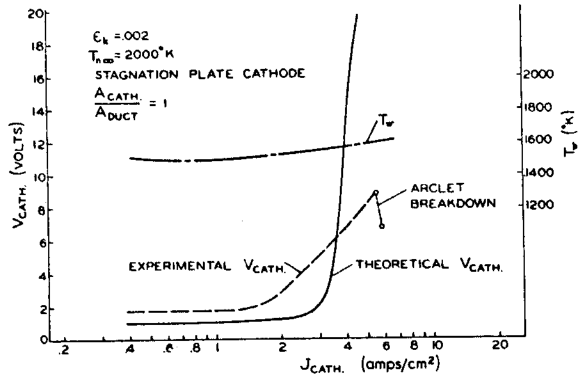

Figure 6. Comparison of Experimental and Analytical Current-Voltage Characteristics.

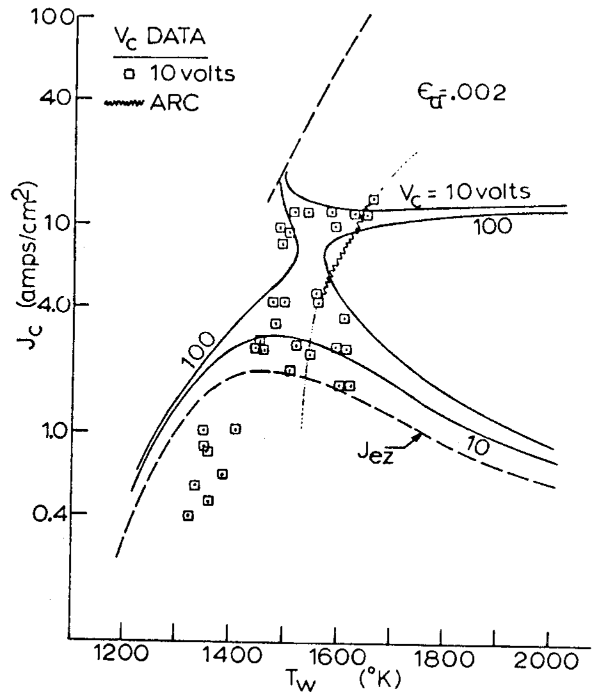

Figure 8. Comparison of Experimental and Theoretical Cathode Voltage Drop Data. Theoretical Results Are Indicated by V_c = const. lines; ϵ_a = .002.

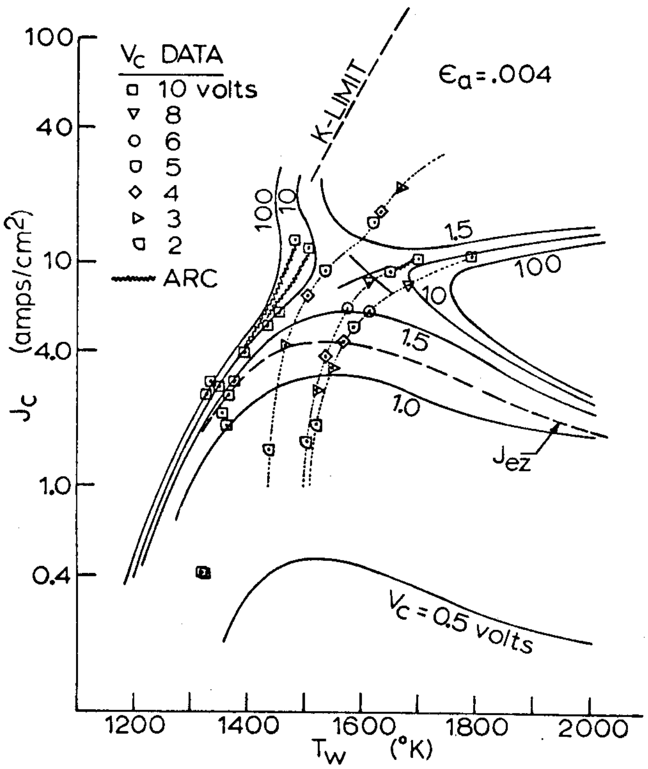

Figure 7. Comparison of Experimental and Theoretical Cathode-Voltage Drop Data. Theoretical Results Indicated by V_c = const. lines; ϵ_a = .004.