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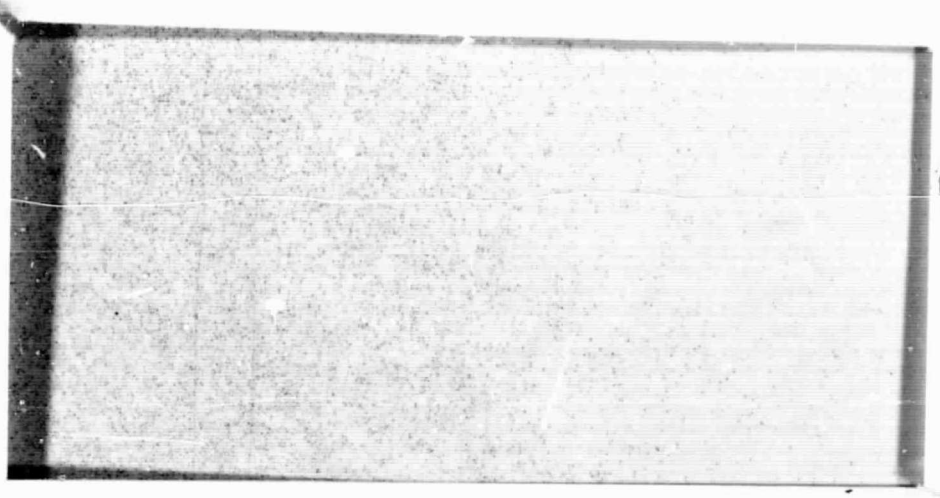

NGL-10-005-089

ENGINEERING AND INDUSTRIAL EXPERIMENT STATION

College of Engineering

University of Florida

Gainesville

Annual Technical Report

on

Experimental Investigations of a Uranium Plasma Pertinent to a Self Sustaining Plasma Source

Written by

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Contract NGR 10-005-089

Reporting Period Jan. 1, 1969 - Dec. 31, 1969

University of Florida

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Gainesville, Florida

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I. INTRODUCTION

This report covers the period from January 1, 1969 through December 31, 1969 of the research done under NASA Grant NGR 10-005-089 entitled "Experimental Investigations of a Uranium Plasma Pertinent to a Self Sustaining Plasma Source."

The following publications describe the results obtained under this NASA Grant.

1. "Measuremet of Temperature and Partial Pressure of a Uranium Plasma," by R. T. Schneider A. G. Randol III, and G. R. Shipman, Applied Spectroscopy, 24, 253-258 (1970).
2. "Temperature Measurement on a Uranium Plasma," by R. T. Schneider, G. R. Shipman and J. M. Mack, Applied Spectroscopy, 23, 671 (1969).
3. "Measurement of the Emission Coefficient of a Uranium Plasma," by R. T. Schneider, A. G. Randol III, C. D. Kylstra and M. J. Ohanian, ANS Transactions, 12, 413 (1969).
4. "Experimental Determination of the Boiling Point of Uranium," by A. G. Randol III, R. T. Schneider and C. D. Kylstra, ANS Transactions, 12, 541 (1969).
5. "Radiation from a Uranium Plasma," by R. T. Schneider, C. D. Kylstra, A. G. Randol III and M. J. Ohanian, ANS Transactions, 12, 3 (1969).
6. "Properties of a Uranium Plasma," by H. D. Campbell, R. T. Schneider and C. D. Kylstra, 8th Aerospace Sciences Meeting, AIAA, New York, Jan. 19-27, 1970.

In the following sections of this report, three papers are reproduced which were given at the Symposium on Research on Uranium Plasmas and Their Technological Applications held on January 7-10, 1970, in Gainesville, Florida. At the time of the writing of this report these papers are not yet available in the literature and they are therefore reproduced in full.

These papers describe the results obtained as of January 1, 1970.

BOILING POINT OF URANIUM

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ABSTRACT

Current techniques for boiling point determinations rely on extrapolation of low temperature vapor pressure data (usually collected below 2500°K) through the Clausius-Clapeyron equation. Since the temperature limitation is, in most cases, difficult to overcome, it is necessary to rely heavily on indirect methods for the evaluation of boiling points. Under special environment conditions there is evidence that an interpretation of uranium plasma electrical characteristics can lead to reasonable estimates of the boiling point.

The device used to generate the uranium plasma, the University of Florida high pressure uranium arc, is described. Details of the electrical power, cooling, high pressure, gas, and data acquisition systems are given.

A modification of the Nottingham equation for electric arc potential versus current is presented. The application of this arc electrical characteristics model is discussed in terms of the determination of the boiling point of the uranium anode. The evaluated boiling points of uranium between 1 atmosphere and 7 atmospheres are compared with the extrapolations of other authors.

THE BOILING POINT OF URANIUM

The experimental uranium plasma program at the University of Florida was initiated in 1966. The successful operation of this first plasma experiment[1] pointed to the potential of further work. A D.C. arc struck between fixed tungsten - uranium electrodes in a sub-atmospheric helium environment generated the plasma. Experimental test times were typically of the order of one to three minutes and were limited by heat dissipation, graphite crucible sublimation, and uranium electrode losses.

The growing interest in the thermal characteristics of uranium in the plasma state prompted a continuation of the program. The extension of the experiment into the regime of gaseous core feasibility resulted in the development[2] of the device shown in Figure 1. As in the prototype, the uranium plasma was generated in a D.C. arc. The system improvements included internal electrode cooling, high pressure capability, a method for arc stabilizing, and an adjustable electrode configuration.

The arc is struck between a cupped tungsten cathode pin and a U-238 anode pellet. The anode pellet sits in a tungsten crucible brazed into the copper anode pedestal. Both of the electrodes are internally cooled with a single-pass water flow. The pressure cell is designed for 200 atmospheres at a wall temperature of 500°F. Cell wall temperatures are held below 150°F with the water cooled, thermal shield.

The chamber pressure environment is derived from a bottled, high pressure gas cylinder. The gas is injected into the chamber in a concentric sheath about the plasma column periphery. The axial down-flow of the gas stabilizes the column during high pressure, high power operation.

Arc ignition is obtained with a pneumatic electrode drive. A manual control initiates the downward motion of the cathode and at the onset of current a sensing device triggers the extraction of the electrode. Electric power is derived from a 40 kilowatt D. C. generator with a maximum current rating of 160 amperes.

In addition to a complete program of plasma temperature and number density diagnostics, the relation between the plasma electrical characteristics and the ambient pressure and temperature was of interest. With an appropriate model it is possible to predict gaseous core engine start-up characteristics assuming that the intiial uranium core volume is to be supplied through a consumable electrode system.

Arc electrical characteristics were analyzed with the model described in Equation 1.

$$V_{ARC} = V_{ANODE} + V_{CATHODE} + \frac{Cdp^m}{I^n} \qquad (1)$$

The voltage drops across the sheath regions of each electrode are accounted for in V_{ANODE} and $V_{CATHODE}$. The term describing the positive column of the arc includes functional dependences on the electrode separation, d, ambient chamber pressure, p, and the arc current, I. The model subjected to computer analysis was

$$V_{OBS} = IR_{SYSTEM} + V_{SHEATH} + \frac{Cdp^m}{I^n} \qquad (2)$$

where an additional term, IR_{SYSTEM}, accounts for voltage drop in the electrodes. The independent laboratory variables were d, p, and I.

A least-squares curve fitting routine was used to determine R_{SYSTEM}, V_{SHEATH}, the constant C, the exponent m, and the exponent n.

Samples of the arc current-voltage characteristics analyzed are shown in Figure 2. The most important characteristic of these curves is the hyperbolic characteristic from which the exponent n of the current I can be evaluated.

The boiling point parameter, n, is related to the boiling point of the anode material through the following expression:

$$n = 2.62 \times 10^{-3} T_{ANODE} \qquad (3)$$

This relationship was the result of work performed and compiled in 1924 by Nottingham[3]. The experiment performed was similar to the experiment in discussion. A subsequent attempt to repeat one of Nottingham's measurements was made by Suits[4]. His failure challenged the value of the arc technique for boiling point determination and has resulted in the use of more direct techniques.

However, under special conditions there is evidence that the Nottingham relationship is reliable. In particular, the following table attests to the fact that an uranium arc in an inert helium atmosphere is well described by the empirical relationship. Table 1 lists the complement of pressures and electrode separations for which the uranium arc current-voltage model was analyzed. The gradual increase of the sheath voltage with increasing pressure is an expected trend and the magnitude indicates that the arc is supported by the uranium species rather than the helium. The computed values of the Nottingham boiling point parameter are listed together with the corresponding evaluation of the anode boiling point. The excellent agreement between the results of this experiment and accepted values for the vapor pressure of uranium is illustrated in Figure 3. The dashed curves are two "state-of-the-art" extrapolations of vapor pressure measurements

made at temperatures below 2100°K. These same measurements were made under vapor pressure conditions 3 orders of magnitude lower than the pressures of interest.

It is important to note that the ordinate is total system pressure, the sum of the uranium and helium partial pressures as measured external to the plasma chamber. The necessary correlation is that the vapor pressure measured in an equilibrium experiment, i.e., effusion experiment, is equivalent to the ambient pressure in the quasi-equilibrium arc method.

Etherington[5] reports an atmospheric boiling point of 4090°K which is within 6% of the value deduced from this experiment.

The early criticism of Nottingham's method was the lack of any theoretical basis. Unfortunately electric arc phenomena is not well understood. However, there is a means available to evaluate the tacit assumption of arc quasi-equilibrium. For the physical picture of anode material evaporating in the plasma column and diffusing radially to the walls of the plasma chamber, it is possible to determine the uranium number density in the arc.

A balance equation for the uranium population in the plasma column can be written as

$$\frac{dN}{dt} = \dot{n} - \beta N \tag{4}$$

where N is the uranium number density in the active plasma volume, $\dot{n}$ is the rate of uranium atom supply, and β is the probability that a particle will remain in the active volume. β is derived from a diffusion concept and is the inverse of the persistence time, τ, which is related to the diffusion coefficient through is related to the diffusion coefficient through the relationship

$$D = \frac{r_o^2}{\tau(2.405)^2} \tag{5}$$

where r_c is the radius of the active plasma volume. τ for this calculation was deduced from the emprical results shown in Figure 4. This work by Raikhbaum and Malykh[6] derived diffusion coefficients from an arc similar to the arc of this experiment. An extrapolatron to a uranium equivalent diffusion coefficient was made.

The uranium number density can then be calculated from

$$N = J \cdot \frac{a}{A} \cdot \tau \qquad (6)$$

under the assumption of quasi-equilibrium

$$\frac{dN}{dt} = 0 \qquad (7)$$

J is the mass flux determined by the temperature of the anode, a Avogadro's number, and A is the uranium atomic mass. The results of this vaporization with diffusion analysis are depicted in Figure 5. In order to relate this calculation to conditions in the plasma column, the effects of ionization must be considered. The number densities calculated are atom densities and in the 8000°K plasma environment the majority of the atoms have been singly ionized.

Figure 6 shows the results of a solution of the Saha Equation at 0.1 atmospheres uranium partial pressure. These results, together with the number density measurements made on the arc[2] indicate that the preceding analysis is close to reality. The vaporization model suggests a number density 20% higher than that measured. However, the accuracy of the uranium partition functions used in the spectroscopic number density measurements is questionable so it is impractical to quote an error limit.

In summary, these authors feel that the arc boiling point technique can be useful in some circumstances. The substantiation of uranium boiling

points beyond the temperature limits of current metallurgy has made a contribution to the uranium technology needed for meaningful gaseous core analysis.

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2. Randol, A. G., III, "A Determination of High Pressure, High Temperature Uranium Plasma Properties," Dissertation, University of Florida, August, 1969.

3. Nottingham, W. B., Physics Review, 28, 1926.

4. Suits, C. G., Physics Review, 55, 1939

5. Etherington, H., Nuclear Engineering Handbook, McGraw-Hill, 1958.

6. Raikhbaum, Y. D., Malykh, V. D., "A Spectroscopic Method for Studying the Diffusion of Atoms on an Electric Arc," Optics and Spectroscopy, Volumn IX, No. 4, October, 1960.

TABLE 1. URANIUM ARC VOLTAGE-CURRENT MODEL

EXPERIMENTAL PARAMETER			COMPUTER RESULTS	
TOTAL PRESS p (Atm)	ELECTRODE GAP d (mm)	SHEATH VOLTAGE V (Volts)	n	TEMPERATURE T (°K)
1.0	4.07	14.28	1.003	3847
2.0	3.0-3.2	16.3	1.0056	3838
2.86	4.75	16.5	1.050	4007
3.72	2.9-3.5	17.9	1.068	4076
4.82	6.01	17.3	1.128	4305
5.6	2.8-3.4	23.06	1.170	4465
6.8	6.01	26.8	1.270	4847

n ≡ NOTTINGHAM PARAMETER

T ≡ ANODE BOILING POINT TEMPERATURE

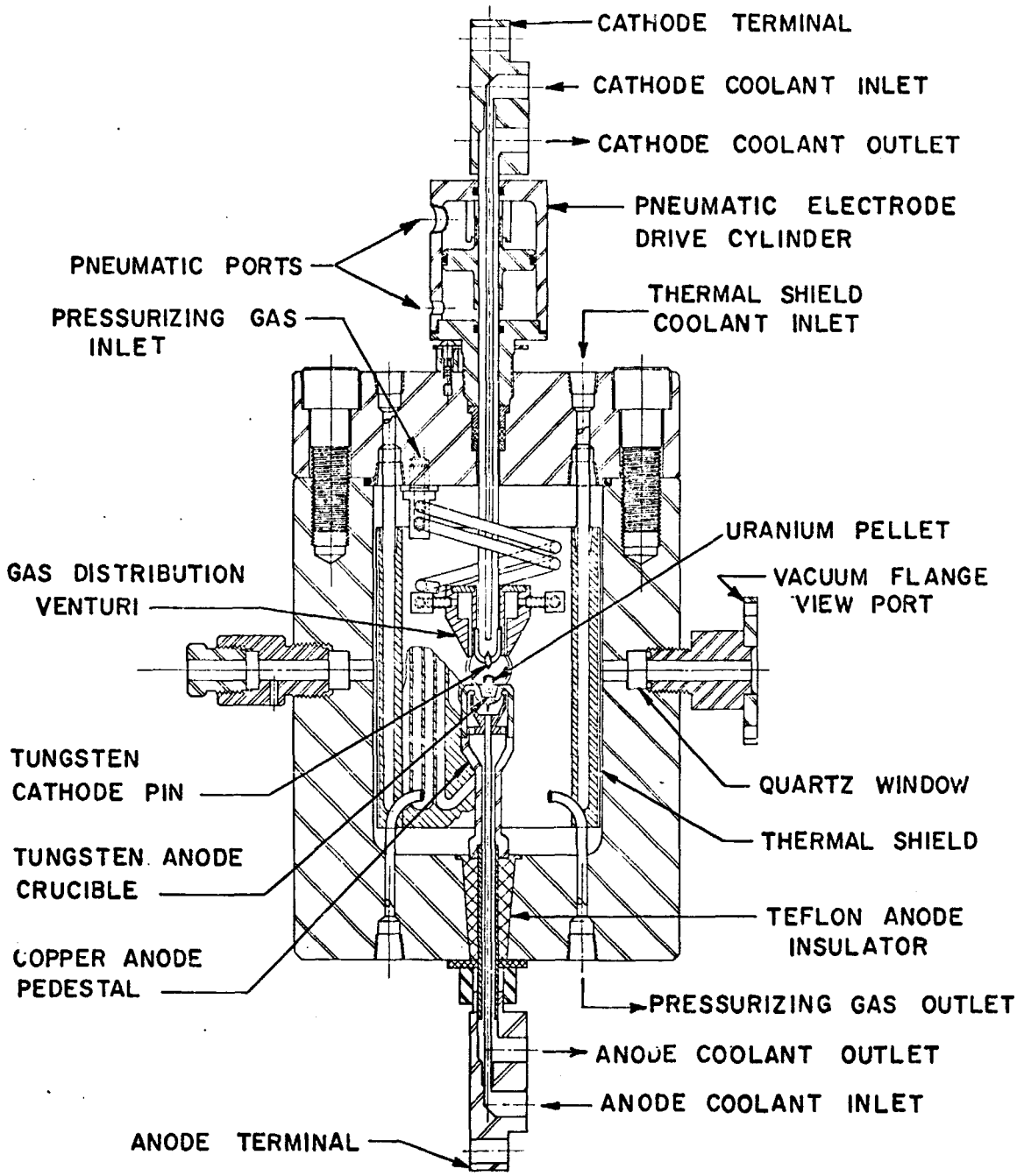

FIG. 1 HIGH PRESSURE URANIUM PLASMA DEVICE

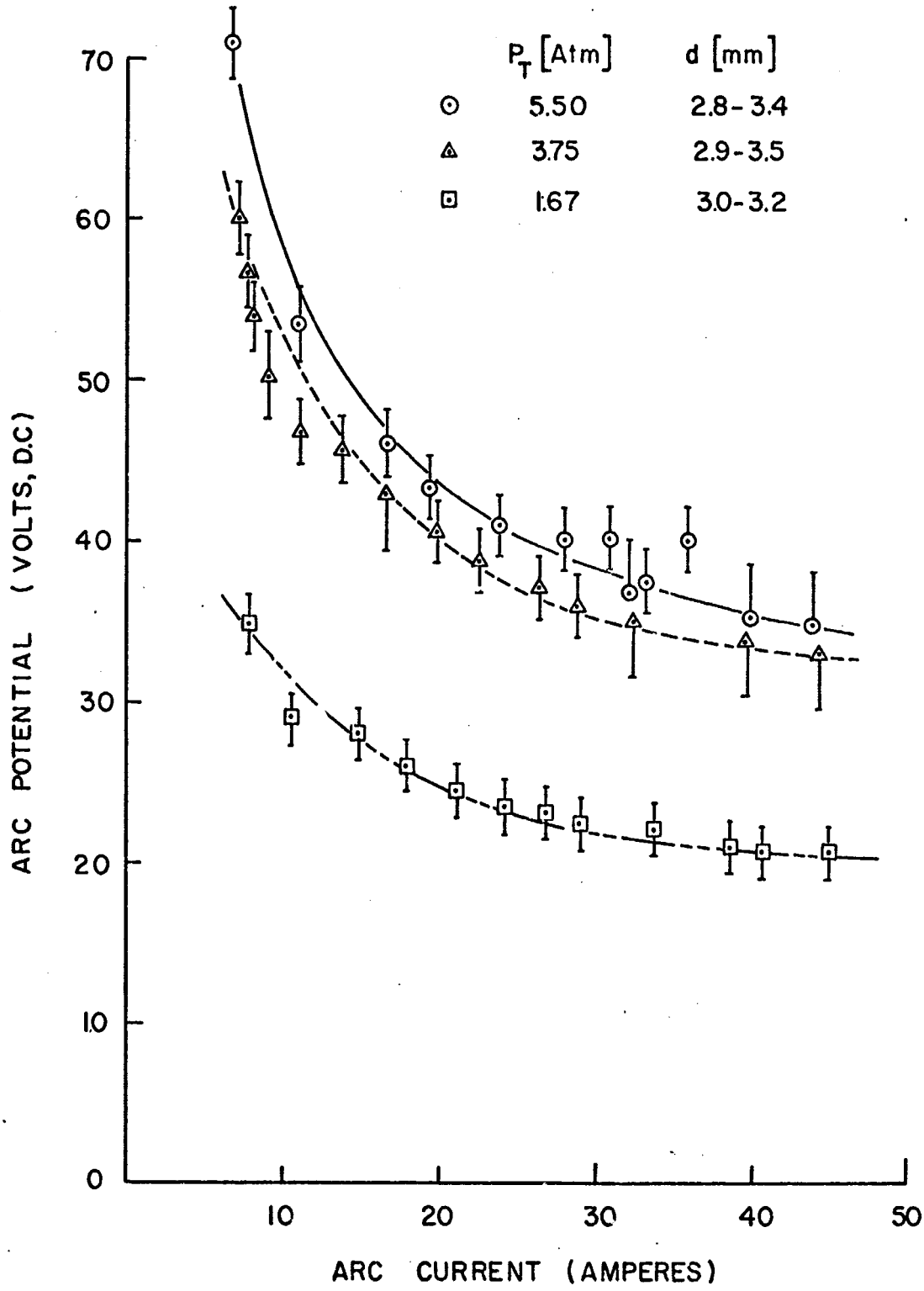

FIG. 2 HIGH PRESSURE URANIUM ARC VOLTAGE-CURRENT CHARACTERISTICS

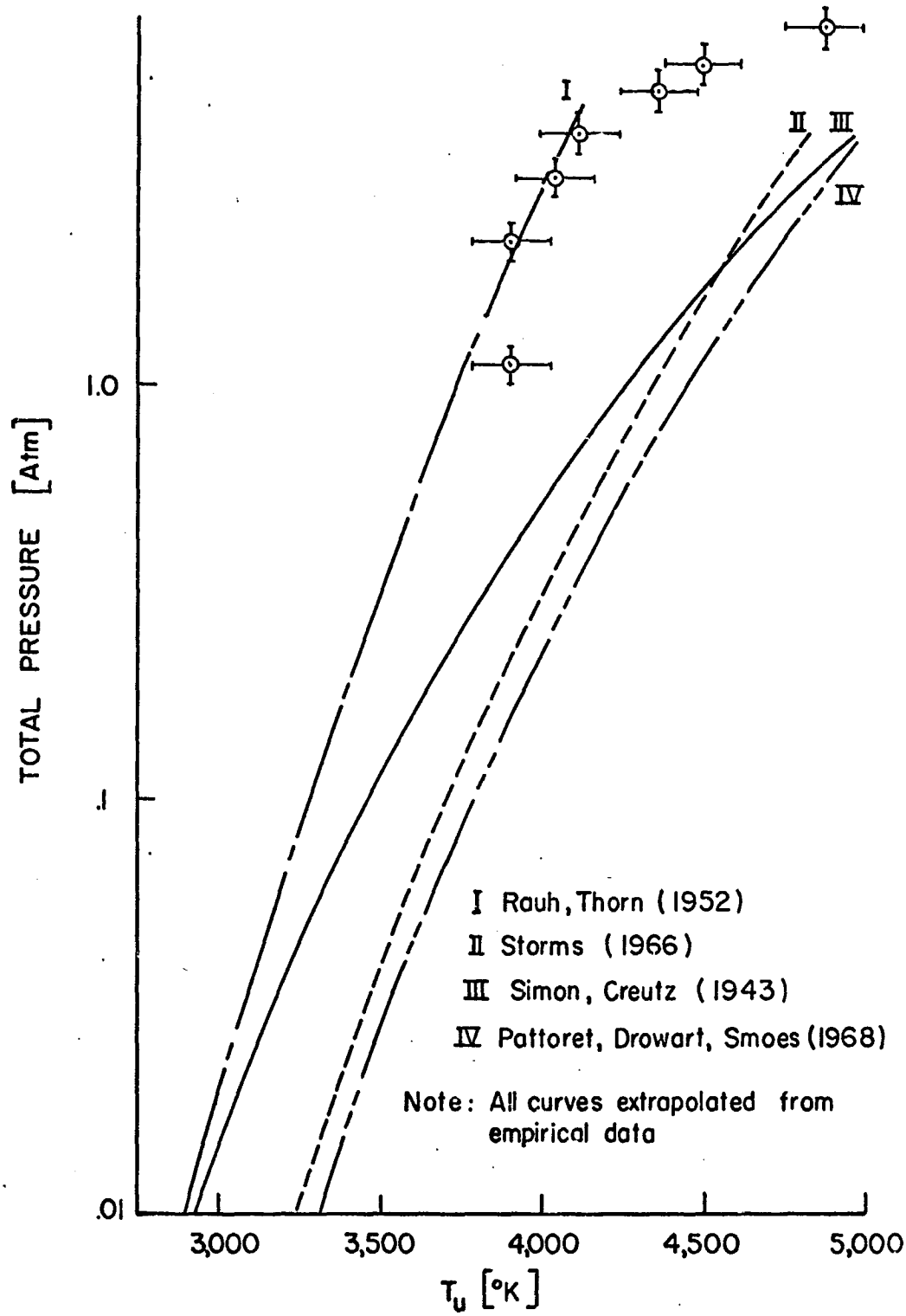

FIG. 3 TOTAL PRESSURE VERSUS ANODE BOILING TEMPERATURE

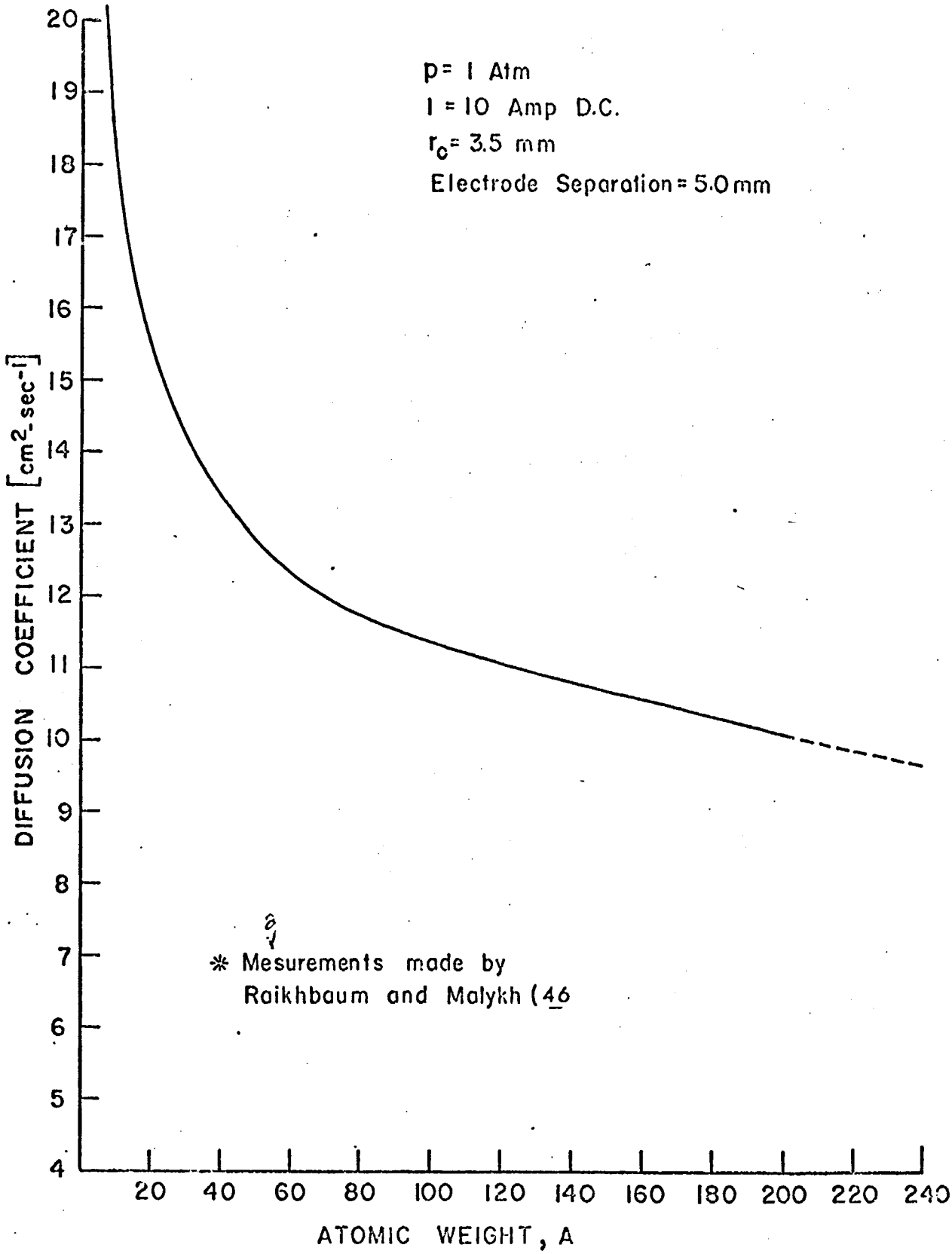

FIG. 4 EXPERIMENTAL* DIFFUSION COEFFICIENTS VERSUS ATOMIC WEIGHT

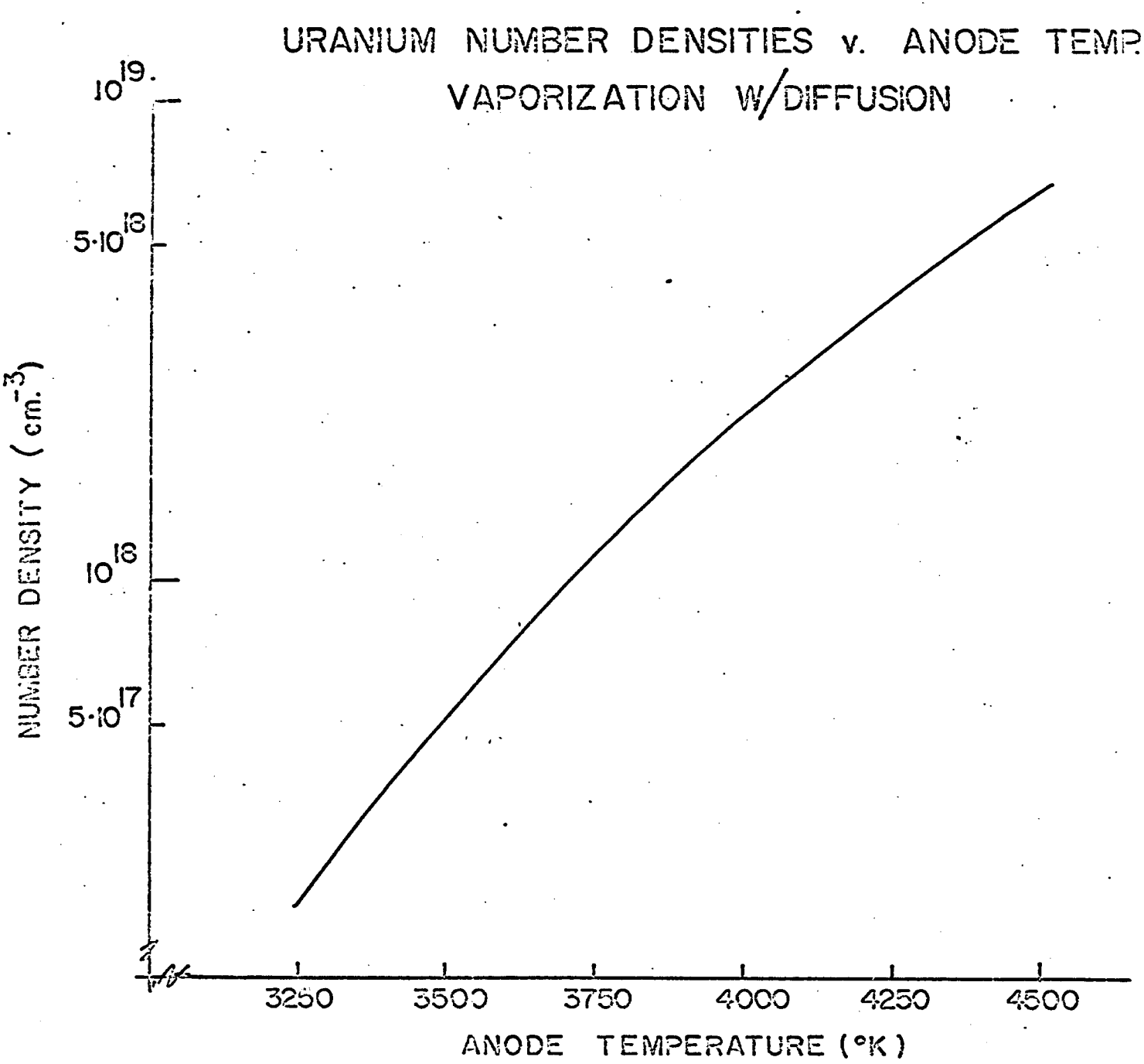
URANIUM NUMBER DENSITIES v. ANODE TEMP.
VAPORIZATION W/DIFFUSION
10^{19}
$5 \cdot 10^{18}$
10^{18}
$5 \cdot 10^{17}$
NUMBER DENSITY ($cm.^{-3}$)
3250
3500
3750
4000
4250
4500
ANODE TEMPERATURE (°K)

FIGURE 5

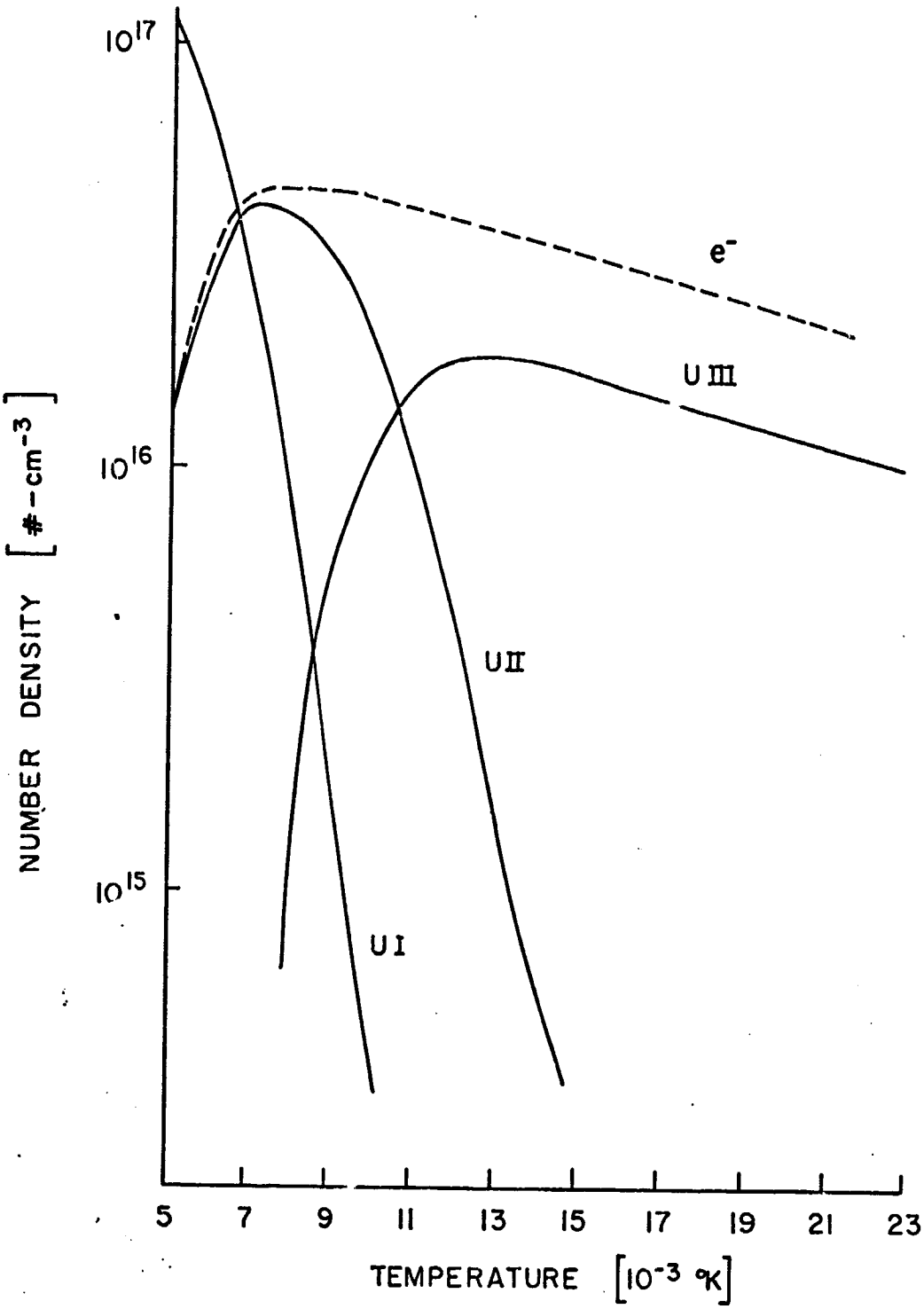

FIG. 6 URANIUM PLASMA COMPOSITION VERSUS TEMPERATURE AT 0I ATMOSPHERE

SPECTROSCOPIC STUDY OF A URANIUM ARC PLASMA*

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ABSTRACT

This paper reports on measurements of temperatures, particle densities, emission and absorption coefficients of a uranium arc. The motivation for this research is to acquire fundamental knowledge of the physical properties of uranium plasmas which will in turn eventually lead to the design of a self-sustaining fissioning plasma.

The arc is generated in a high pressure cell capable of withstanding pressures up to 100 atmospheres. The current passes between a tungsten pin cathode and a uranium pellet anode. A cover gas of helium is used and substantial vaporization of uranium takes place as the pellet temperature rises and the arc stabilizes. The atomic characteristics of the helium and uranium are such that the emitted line radiation originates primarily from singly-ionized uranium.

Spectroscopic diagnostics were used throughout the investigation. Several methods of temperature determination were employed including Boltzmann plots, relative line intensities and brightness emissivity methods. Although there are difficulties associated with each method all temperature measurements were found to lie in the range of 7500-12,000°K.

The measurements of emission and absorption coefficients of the plasma were taken over the pressure range of 3-15 atmospheres and current range of approximately 15-50 amperes. The corresponding partial pressure of uranium over this range was estimated to be 0.1 - 0.5 atmospheres.

INTRODUCTION

Interest in uranium plasmas has arisen from the possibility of using a self-sustaining uranium plasma as a plasma

*Work supported by NASA, Contract NRG-10-005-089.

source - the gaseous core reactor. Because of the very high temperatures associated with such a device, it will be necessary to remove the energy generated in the reactor by radiative coupling to some working fluid. Hence, it is necessary to study the optical and kinetic properties of high pressure uranium plasmas.

Of particular interest for future design considerations would be knowledge of emissive power as a function of operating pressures, temperatures, wavelengths, and system geometry. Calculations concerning criticality, radiative heat transfer to the working fluid, and system startup and control all require the above information. At present, these calculations must rest upon theoretical predictions. Unfortunately, however, the basic constants and properties of uranium which are required for such predictions are poorly known.

The need for experimental work on uranium plasmas to measure radiation emission characteristics is clearly indicated. This paper describes some of the initial spectroscopic measurements on a uranium arc plasma, and the diagnostic methods employed to analyze the data. The plasma device has been described in the preceding paper of these proceedings.

MEASUREMENTS OF TEMPERATURE AND PARTIAL PRESSURES

Spectroscopic diagnostics - both photographic and photoelectric - were used throughout the investigation. The basic requirement for evaluation of spectroscopic measurements discussed in this section is the achievement of local thermal equilibrium (LTE) within the plasma. The different criteria for establishing the existence of LTE (ref. 1) were examined, and it was shown that the LTE assumption was valid for the plasma conditions under which data were taken.

Uranium Spectrum

The first experimental investigation of uranium spectra was undertaken in 1946 (ref. 2) and until recently there has been no serious effort to catalog any data beyond the first ionization stage. The results reported by Corliss and Bozmann (ref. 3) were generated by an electric arc operating at approximately 5,000°K. At this temperature no significant radiation from the doubly or higher ionized species is expected.

In the present experiment, the task of evaluating the emitted uranium spectra was not as difficult as in the one

mentioned above (ref. 3). Both the arc temperature and the uranium particle density were relatively high. Consequently, the majority of observed emission lines were those characteristic of singly ionized uranium (UII). In addition, only the strongest UI (atomic uranium) and a trace of helium lines were observed (ref. 4). From this spectrum evaluation it was possible to estimate the maximum arc temperature to be within the range 8,000°K to 15,000°K. Preliminary temperature measurements (ref. 5) verified this to be so.

The uranium atom naturally possesses a complicated electronic structure as well as a highly complex emission spectrum. So far, no fewer than 1159 UI lines and 315 UII lines have been catalogued (ref. 6) between 2000 Å and 9000 Å. This high density of spectral lines complicates standard diagnostic techniques, and suitable modifications of standard methods is required before reliable measurements can be performed.

The Boltzmann Plot

The Boltzmann Plot method is a relative line method which is independent of number densities and partition functions.

The emission coefficient, ε, for line radiation between two excited states (i.e. power radiated per unit volume per unit solid angle) is given by

$$\varepsilon = \frac{h\nu}{4\pi} A\, n\, \frac{g}{Z} \exp\,(-E/kT) \qquad (1)$$

where $h\nu$ is the photon energy of the transition; A is the transition probability; n is the total population density of the species; g is the statistical weight of the upper level of the transition; Z is the partition function of the species; E is the energy of the upper state; k is Boltzmann's constant; and T is the temperature. This relation is valid if LTE is established within the plasma. The intensity (I) of emitted radiation for an optically thin plasma is:

$$I = \int \varepsilon dl = \bar{\varepsilon}\, L \qquad (2)$$

where L is the plasma thickness along the line of sight and $\bar{\varepsilon}$ is the average value of the emission coefficient. Using the preceding two equations, plus the relation between wavelength and frequency ($\lambda = c/\nu$), and taking the logarithm of both sides of the equation, we obtain

$$\log \frac{I\lambda}{gA} = C - \frac{5040\ E}{T} \qquad (3)$$

where E and T are expressed in eV and ^{o}K respectively. Now T is some average temperature along the line of sight. Normally, it is within 10% of the maximum temperature. This equation is that of a straight line where the slope can be related to the temperature.

When such a temperature graph (Boltzmann Plot) is constructed for different values of log [Iλ/gA] measured from an arc, the points may not fall on a straight line for several reasons. Self-absorption and inhomogeneity along the line of sight, as well as experimental errors will result in nonlinearity. These phenomena are common since the light emitted from the high-temperature region of the arc center must traverse the low-temperature region at the edge of the arc, where the lower levels are relatively much more populated with atoms capable of absorbing. The plotted points usually have an upper envelope which consists, to a good approximation, of a straight line of negative slope; this straight line is the locus of those points which pertain to spectrum lines emitted under conditions of optical thinness (ref. 7).

If it is possible to determine by some independent means, such as through an absorption experiment, that the plasma is indeed optically thin for a majority of the spectral lines selected for the Boltzmann Plot, the slope may be determined by a least squares fit.

Figure 1 shows a Boltzmann Plot of arc centerline intensities used to characterize the plasma temperature as a function of pressure. The curves shown are the linear least squares fit to the data points. The temperature is then determined from the slope of these curves. The increase in temperature for increasing pressure is expected since at constant current the power input increases.

Figure 2 shows a family of plasma temperature profiles which were derived from Boltzmann Plot temperatures at the chordal position (x/X) corresponding to the radial positions (r/R).

The Fowler-Milne Method

This method relies on the fact that the emission coefficient for any given spectral line passes through a maximum at a unique temperature. It is possible to determine this "norm-temperature" (ref. 8) by solving

$$\frac{d\epsilon}{dT} = 0 \qquad (4)$$

for the unique temperature T_n. The solution of this equation is coupled to the pressure through the ground state number density evaluated with Saha's equation (see equation 7).

For the experiment reported here, the radial emission coefficient profile exhibits an off-axis maximum for each line studied. The method of analysis thus requires that a value be assigned to the norm-temperature T_n, which is a function of the uranium partial pressure. One can then proceed to solve the relative line relationship

$$\frac{I(r)}{I(r_n)} = \frac{Z(T_n)}{Z(T_r)} \cdot \frac{n(r)}{n(r_n)} \cdot \exp \frac{Eu}{k}\left\{ \frac{1}{T_r} - \frac{1}{T_n} \right\} \qquad (5)$$

for the temperature profile T(r), corresponding to the observed intensity, I(r).

Calculation of Emission Coefficients

Referring to equations 1 and 2 it can be seen that the intensity of an emission line is temperature dependent in a manner dictated by the temperature dependence of the three quantities, n(T), Z(T), and exp (-E/kT). Although the last quantity is easily handled the first two are not. The techniques and approximations required for determining the form of n(T) and Z(T) will now be discussed.

Assuming the plasma to be in LTE, then the following form of the ideal gas law is a good equation of state:

$$P = n_t kT = kT\sum_i n_i \qquad (6)$$

where P is the total pressure; n_t is the total particle density; and n_i is the particle density of the i^{th} species (i.e. electrons, neutrals, ions, etc.)

For a given temperature Saha's equation can be used to relate the densities of particles in different stages of ionization:

$$\frac{n_{i+1} n_e}{n_i} = \frac{2Z_{i+1}(T)}{Z_i(T)} \cdot \left\{ \frac{2\pi m_e kT}{h^2} \right\}^{3/2} \cdot \exp -\left\{ \frac{IP-\Delta E}{kT} \right\} \qquad (7)$$

where n_i is the particle density of the i^{th} ionized species; IP is the ionization potential of the i^{th} species; and ΔE is lowering of the IP due to electric microfields in the plasma (ref. 9).

One additional relation is required in order to uniquely determine the particle densities and that is the assumption of charge neutrality. Charge balance is expressed by:

$$n_e = \sum Z_i n_i \qquad (8)$$

where Z_i is the positive ion charge.

The above equations can now be solved to give the plasma composition as a function of temperature and total pressure. Writing Saha's equation in the form

$$\frac{n_{i+1} n_e}{n_i} = S_{i+1}(T) \tag{9}$$

and using equations 6, 7, and 8 we obtain the following cubic equation in n_e:

$$n_e^3 + (2S_2) n_e^2 + (3S_1S_2 - PS_1/kT)\, n_e - 2PS_1S_2/kT = 0 \tag{10}$$

This expression assumes that only singly and doubly ionized uranium ions are present (as indicated previously). The single positive root of the above equation is the desired solution for n_e. This solution then leads to a complete description of the plasma composition. Number densities for a pressure of 0.1 atmospheres are shown in figure 3. Since the third ion stage is not included in these calculations, the UIII and electron number densities shown are only approximate for temperatures greater than 15,000°K. The predominant species at typical uranium arc temperatures is the first ion, hence the lowering of the ionization potential, which is proportional to z_i^2, can be neglected in the Saha equation solution.

In the plasma column the total pressure is determined by the sum of the helium partial pressure, the electron pressure, and the sum of the uranium species partial pressures. Because of the presence of helium, it would appear necessary to include the helium ionization equation in the above calculations. However, the high ionization potential of helium inhibits any significant ionization below 15,000°K.

It has been tacitly assumed so far that the partition functions are known. Unfortunately this is not so. There appears to be disagreement between reported theoretical values (ref. 10 and 11). In order to proceed it was necessary to evaluate the available information and construct a consistent set of partition functions.

By definition, the partition function of the i^{th} species is a sum over all available energy levels:

$$Z_i(T) = \sum_u g_{iu} \exp\left\{ -E_{iu}/kT \right\} \tag{11}$$

where g_{iu} and E_{iu} are the statistical weights and energies of the uth excited energy level of the i^{th} species. At low temperatures one can approximate the partition function by the statistical weight of the ground state. For UI the

ground state value is 85 and for UII this value is 52.

Figure 6 shows available values for the partition functions of UI, UII, and UIII. The curves of reference 10 were anchored to the values of 52 for UI and 82 for UII for the ground states. Figure 4 is a modification of these curves using the correct ground state degeneracies. The values computed by Waber (ref. 11) for UII were in good agreement up to approximately 10,000°K.

A two-term exponential function was used to approximate these values (least squares fit) to provide simple temperature dependent expressions. The results of the fit were:

$$Z_{UI} = 46.25 \exp(.159T) + 755.2 \exp(-1.69T)$$

$$Z_{UII} = 9.201 \exp(.21T) + 35.603 \exp(.05T) \qquad (12)$$

for temperature T, in units of 10^{-3}°K. These fitted functions were accurate to within 5% of the reported values from 5000°K to 15,000°K.

Referring back to equation 1, it is evident that it is now possible to determine the temperature dependence of the emission coefficient. Figure 5 shows the results of these calculations for several pressures.

Temperature Profiles from Arc Intensity Profiles

The radial analysis of a spectral line, using a density gradient corresponding to a radial plasma intensity gradient, yields an arc intensity profile. Two of these profiles are shown in figure 6. One profile represents the intensity distribution before Voigt analysis (ref. 13) and the other represents the distribution after Voigt analysis. The pronounced difference between these profiles is observed in zones near the arc centerline where the pressure is highest and pressure broadening is dominant. The data points extend to only zone 10 because photographic analysis of the spectral line beyond that point is impractical due to low radiation intensity. The temperature profile of the arc can be obtained from the intensity profile (figure 6) by using the results of the line intensity calculations (figure 5). However, knowledge of the number densities of uranium is required.

Partial Pressure of Uranium

The number densities can be obtained in the following manner. A set of uranium pressures varying between 0.1 - 3.0 atm is assumed and an intensity profile as shown in figure 6

is evaluated using the assumed pressures. This results in a family of temperature profiles for one single measured intensity profile, the assumed pressure being the parameter as indicated in figure 7. Since these intensity profiles have to agree with the temperature profile obtained by the Boltzmann Plot method, it is now possible to select the appropriate uranium pressure for each location in the arc. Wherever the Boltzmann Plot profile in figure 6 intersects one of the relative intensity profiles the pressure parameter attached to the intersected curves is the correct uranium pressure.

The final result is given in figure 8 for two different operating conditions of the arc. Large uncertainties are introduced into this measurement due to the errors propagated through the Abel inversion of the intensity profiles. No realistic error can be assigned, since part of the error is due to the assumption that one temperature is sufficient to describe the emission from a given zone. Another difficulty encountered is the high sensitivity of the pressure determination to errors in the temperature.

It can be assumed that the partial pressure given in figure 8 for the center of the arc is of reasonable accuracy. Due to the fact explained above the accuracy decreases when going to the outer zones of the arc. The "pressure walls" indicated in figure 8 are certainly real, however, their absolute height may be exagerated for reasons discussed in the next chapter.

MEASUREMENT OF ABSORPTION AND EMISSION COEFFICIENTS

The "Brightness Emissivity Method" (or line reversal method) is generally applied to homogeneous transient plasmas such as those produced in shock tubes (ref. 12). In fact, the plasma should be in thermal equilibrium and uniform in order for the method to be applicable. Although the uranium plasma exhibits LTE it is definitely not uniform. Consequently, only spatially averaged quantities can be measured.

The underlying theory and practical applications of the method are clearly described in the literature (ref. 8, 12), and so only the important features are presented here.

Essentially one proceeds by measuring the intensity of radiation emitted by a plasma both with and without a background standard source. The absorption of the background source radiation by the plasma can be used to determine both the temperature and optical thickness of the plasma. Measurements have to be taken at a location in the spectrum where the plasma is radiating a strong emission line in order that significant absorption takes place.

The equation of radiative transfer

$$\frac{dI}{dx} = \epsilon - \kappa I \tag{13}$$

where κ is the absorption coefficient, has been solved for simple geometries. In particular, if the intensity of the plasma and background source alone are Ip and I_F respectively, and if the intensity of the plasma and background source together is I_T, then

$$I_T = I_P + I_F e^{-\tau} \tag{14}$$

where τ is the optical thickness of the plasma at that wavelength. If the thickness of the emitting plasma is L, then the average absorption coefficient $\bar{\kappa}$ is given by

$$\bar{\kappa} = \frac{\tau}{L} = \frac{1}{L} \log\left\{\frac{I_F}{I_T - I_P}\right\} \tag{15}$$

If Wien's function is used to approximate the blackbody Planck function then the following relation can be derived

$$\frac{1}{T_P} = \frac{1}{T_F} + \frac{\lambda}{C_2} \log\left\{\frac{I_F + I_P - I_T}{I_P}\right\} \tag{16}$$

where T_P, T_F are the temperatures of the plasma and background source respectively; λ is the wavelength at which the measurement is being taken; C_2 is a constant. The background source (a high intensity flash lamp) was calibrated with both a tungsten filament lamp and a carbon arc.

Figure 11 displays the measurements of the absorption coefficient for the UII 4090 Å emission line as a function of pressure. The average temperature of the plasma for these measurements was only 7500°K, a value slightly lower than mentioned previously.

These results indicate that the line optical thickness is much higher than might have been expected when interpreting the temperature measurements from the Boltzmann Plot (see figure 1). The effect of this high optical thickness would be an overestimate of the Boltzmann Plot temperature, which would in turn result in an overestimate of the uranium partial pressures (see figure 8) and in particular, an overestimate of the "pressure walls."

The average line emission coefficient can be determined from the previous measurements by using Kirchoff's Law. Of more practical value however is information of the radiation emitted over the entire spectrum.

A thermopile and a series of optical filters were used to measure the emission coefficient over the visible spectrum (approximately 3300 Å - 7000 Å). Various combinations of filters were used to obtain data in approximately 700 Å step intervals. The measurements represent averages along the line of sight taken at the center of the arc. Typical results for two sets of operating conditions are shown in figure 10.

CONCLUSIONS

The results of our research program to date have been primarily directed towards development of diagnostic techniques for measuring properties of a uranium plasma. A range of different approaches has been taken, all with varying degrees of success, but on the other hand, all yielding reasonably similar results.

The objective of the work was to obtain measurements of emission and absorption coefficients of a uranium plasma as a function of pressure and temperature. This goal has been partially achieved in that a number of measurements directed to this end have been obtained. The results reported here are typical of a uranium plasma of pressure .1 - .5 atmospheres at a temperature of 7500 - 10,000°K.

In conclusion it should be said that the measurements are of a preliminary nature. In order to cover a sufficient pressure and temperature range - which is essential for plasma core reactor design - further research is required.

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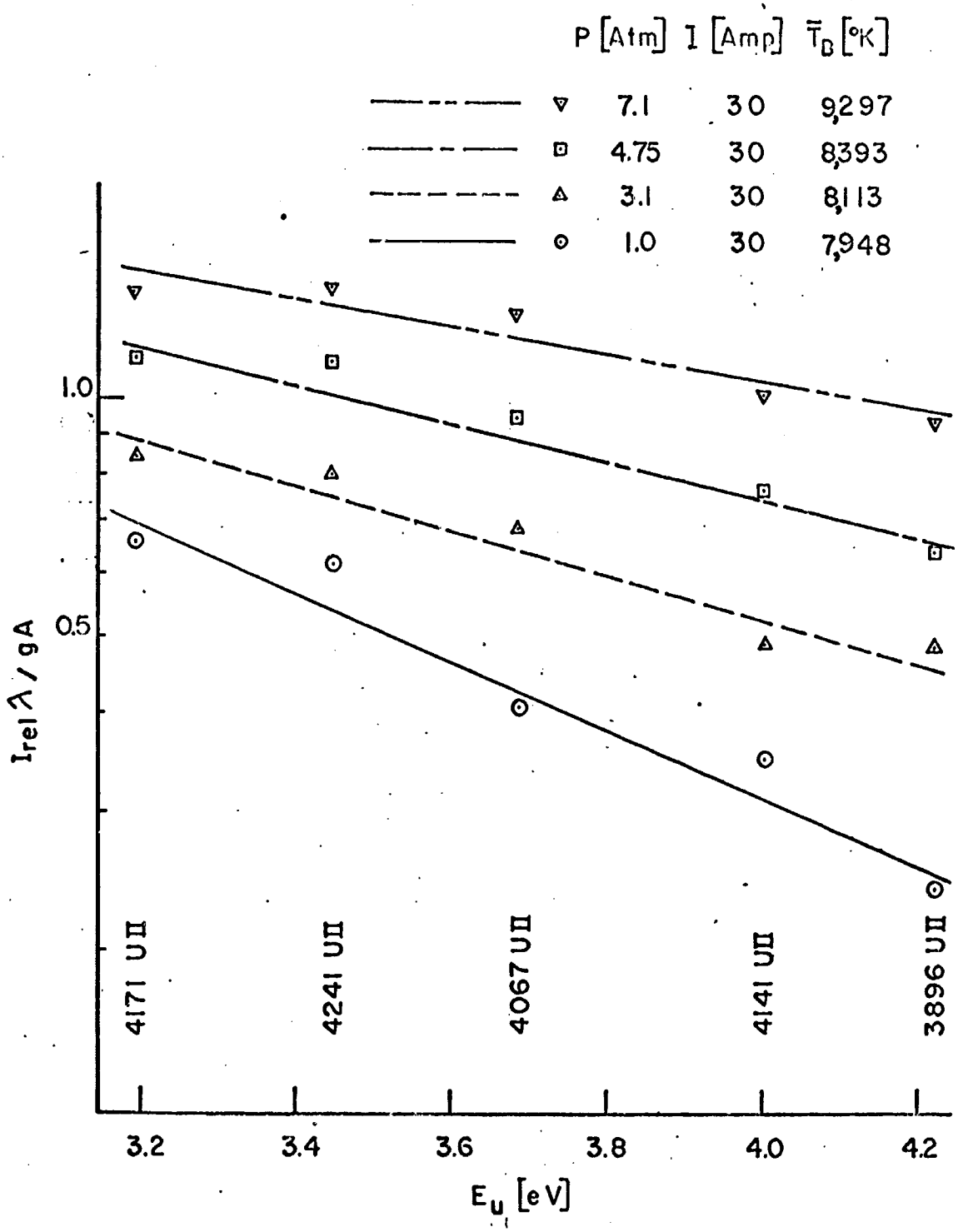

FIG. I IONIC BOLTZMANN PLOT USING CHORDAL INTENSITIES OF UII LINES

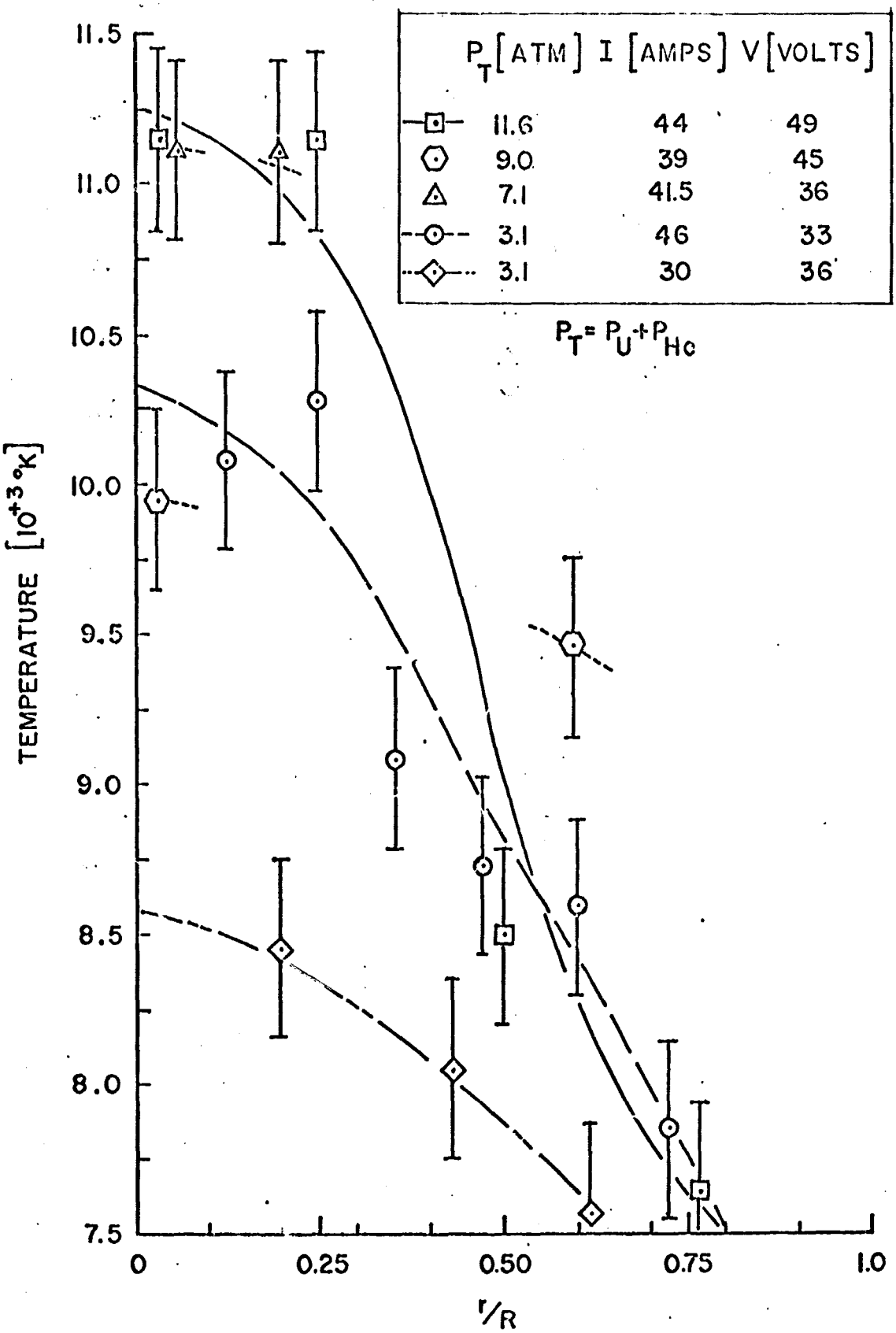

FIG. 2 URANIUM PLASMA TEMPERATURE PROFILES FROM BOLTZMANN PLOT DETERMINATIONS

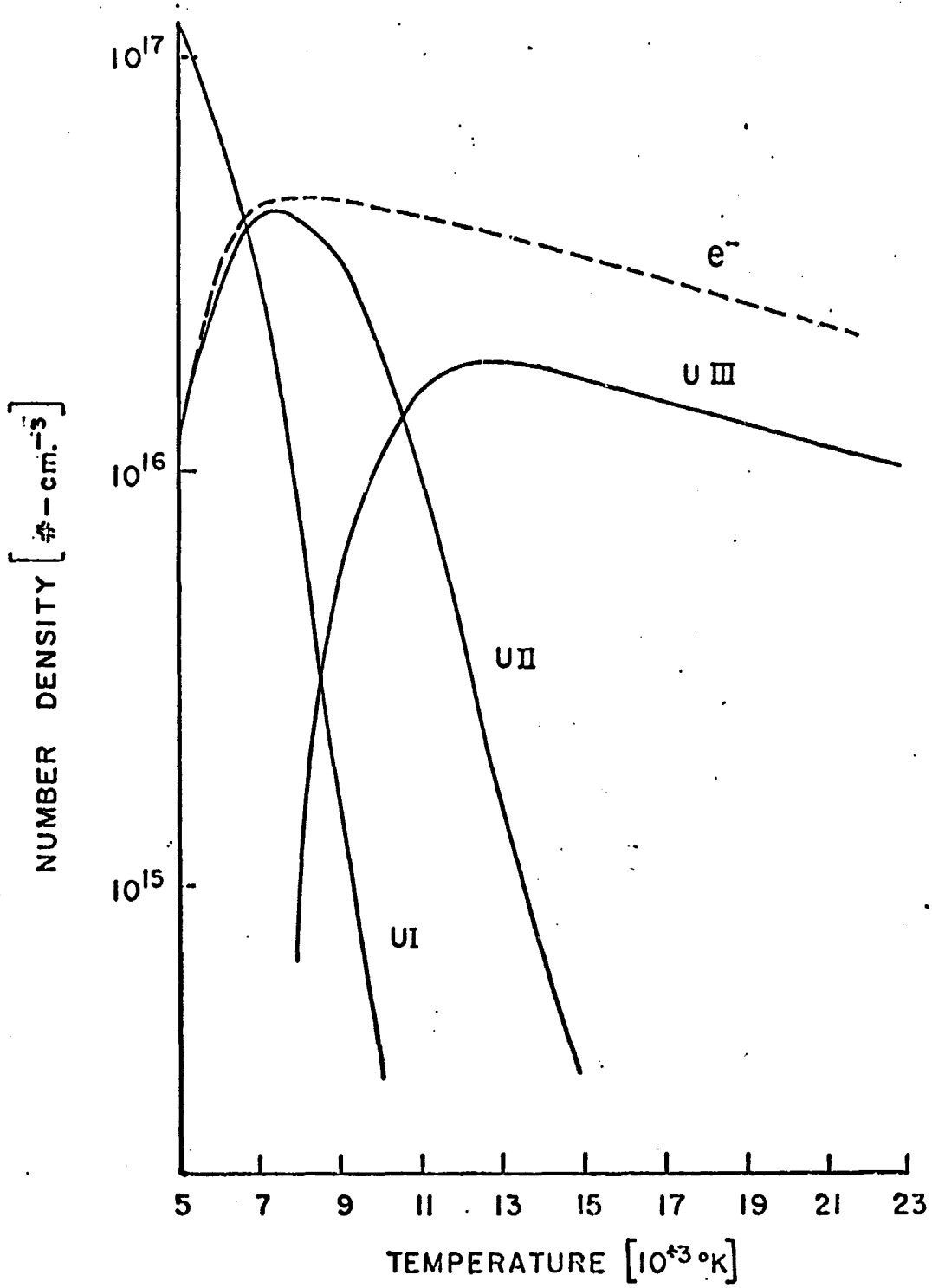

FIG. 3 URANIUM PLASMA COMPOSITION VERSUS TEMPERATURE AT 0.1 ATMOSPHERE

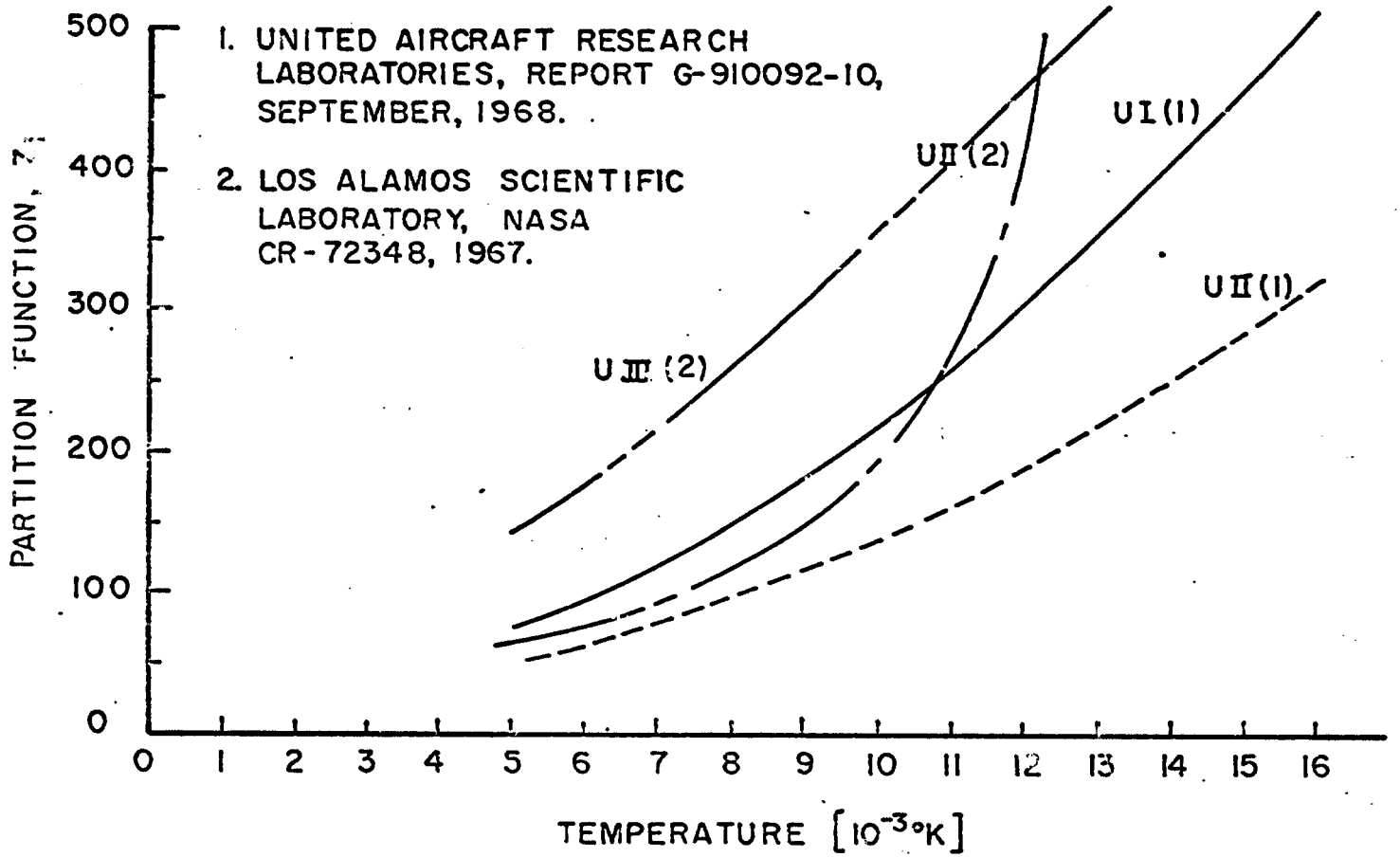

FIG. 4 TEMPERATURE DEPENDENCE OF URANIUM I, URANIUM II, AND URANIUM III PARTITION FUNCTIONS

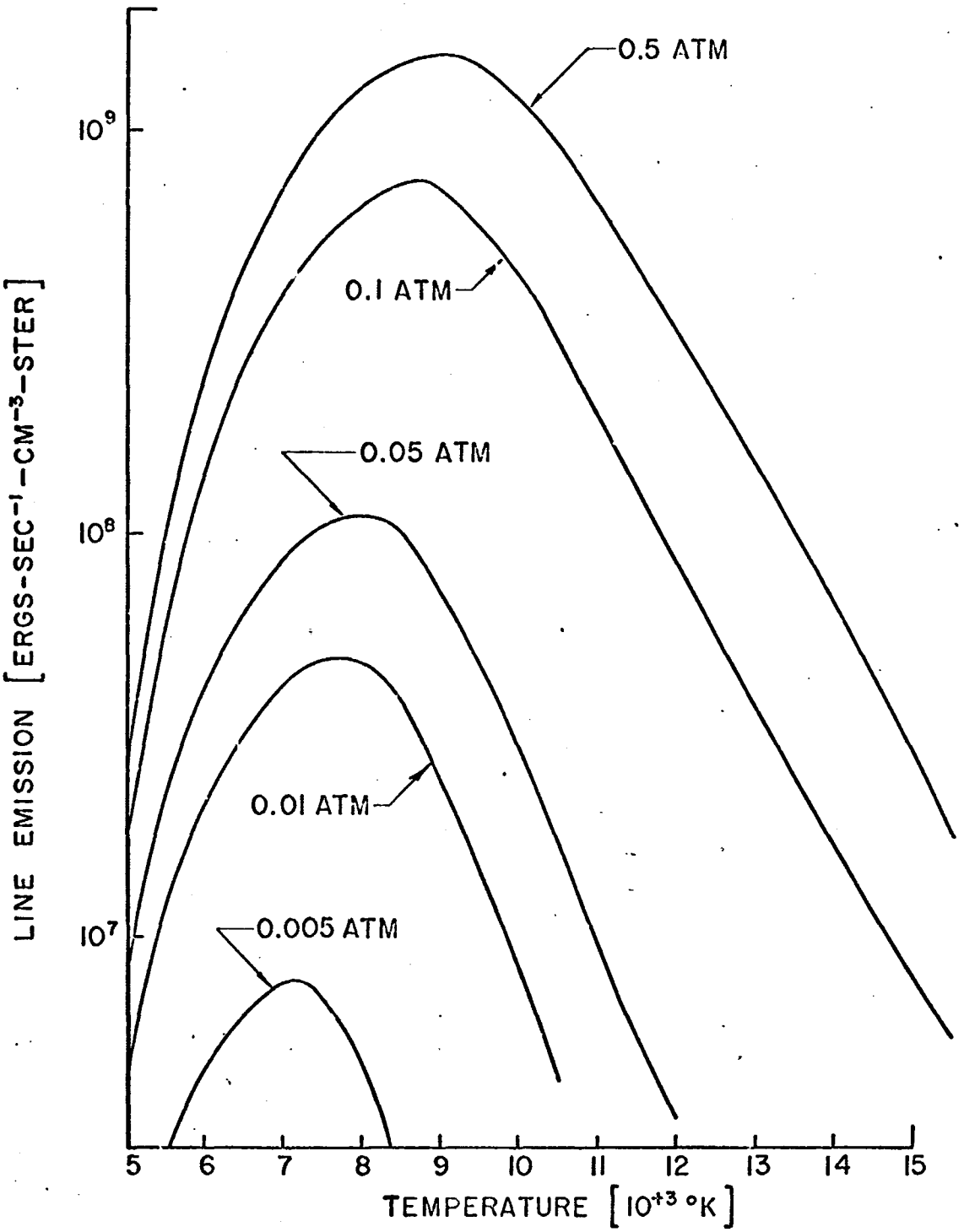

FIG. 5 LINE EMISSION OR THE 4171 UII TRANSITION VERSUS TEMPERATURE FOR DIFFERENT PRESSURES

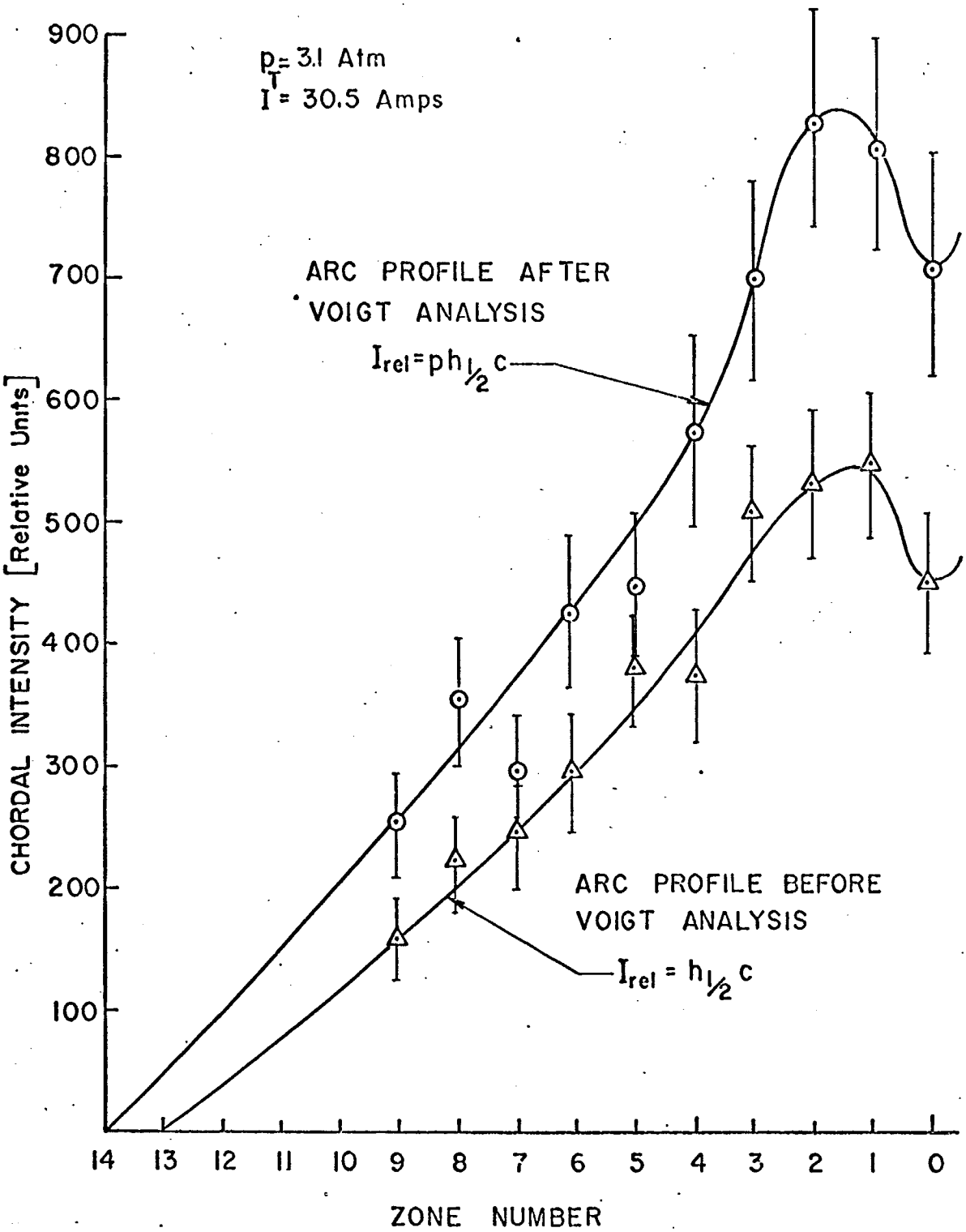

FIG 6 ARC INTENSITY PROFILES WITH AND WITHOUT VOIGT ANALYSIS

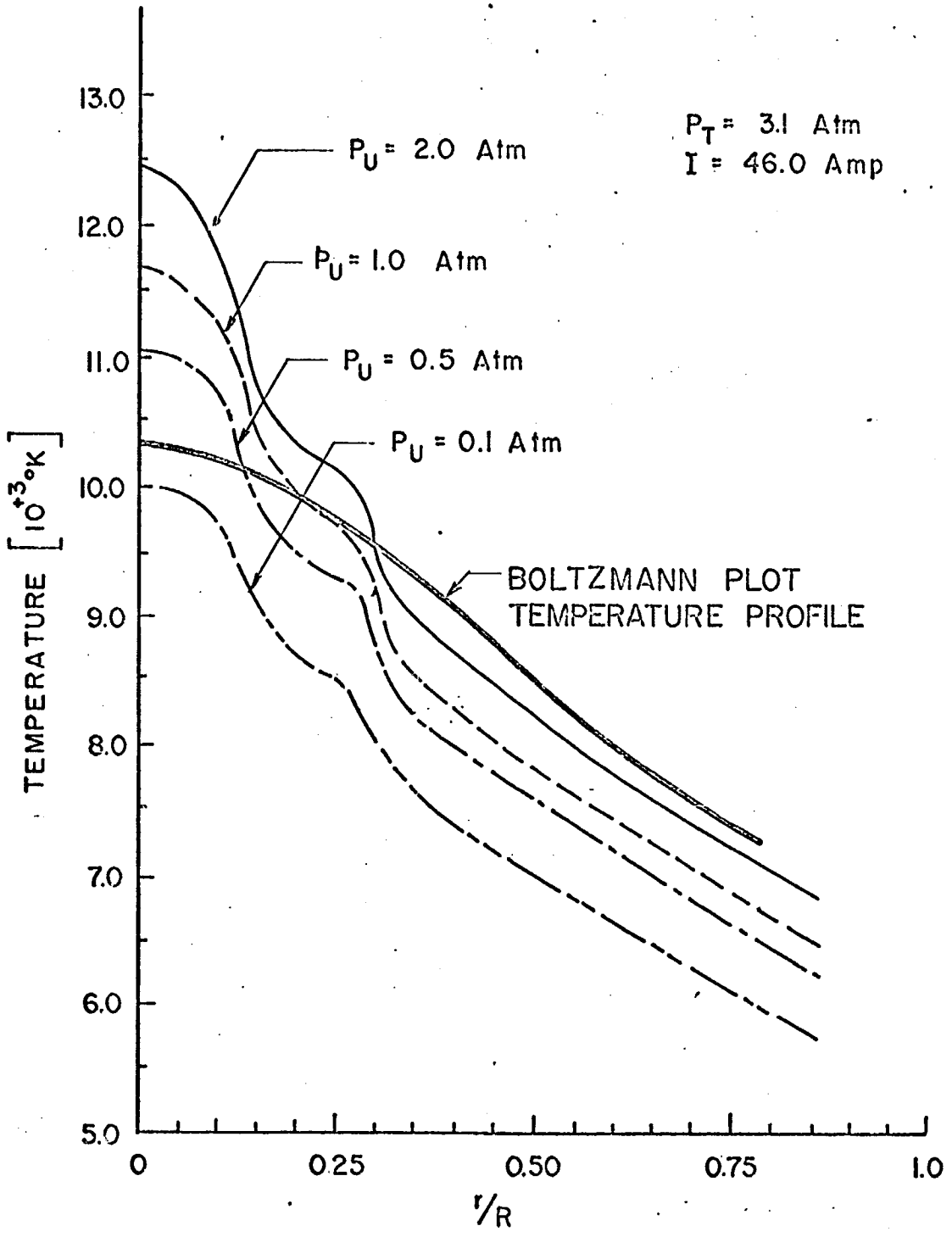

FIG. 7 URANIUM PLASMA TEMPERATURE PROFILES FROM RELATIVE INTENSITY ANALYSIS

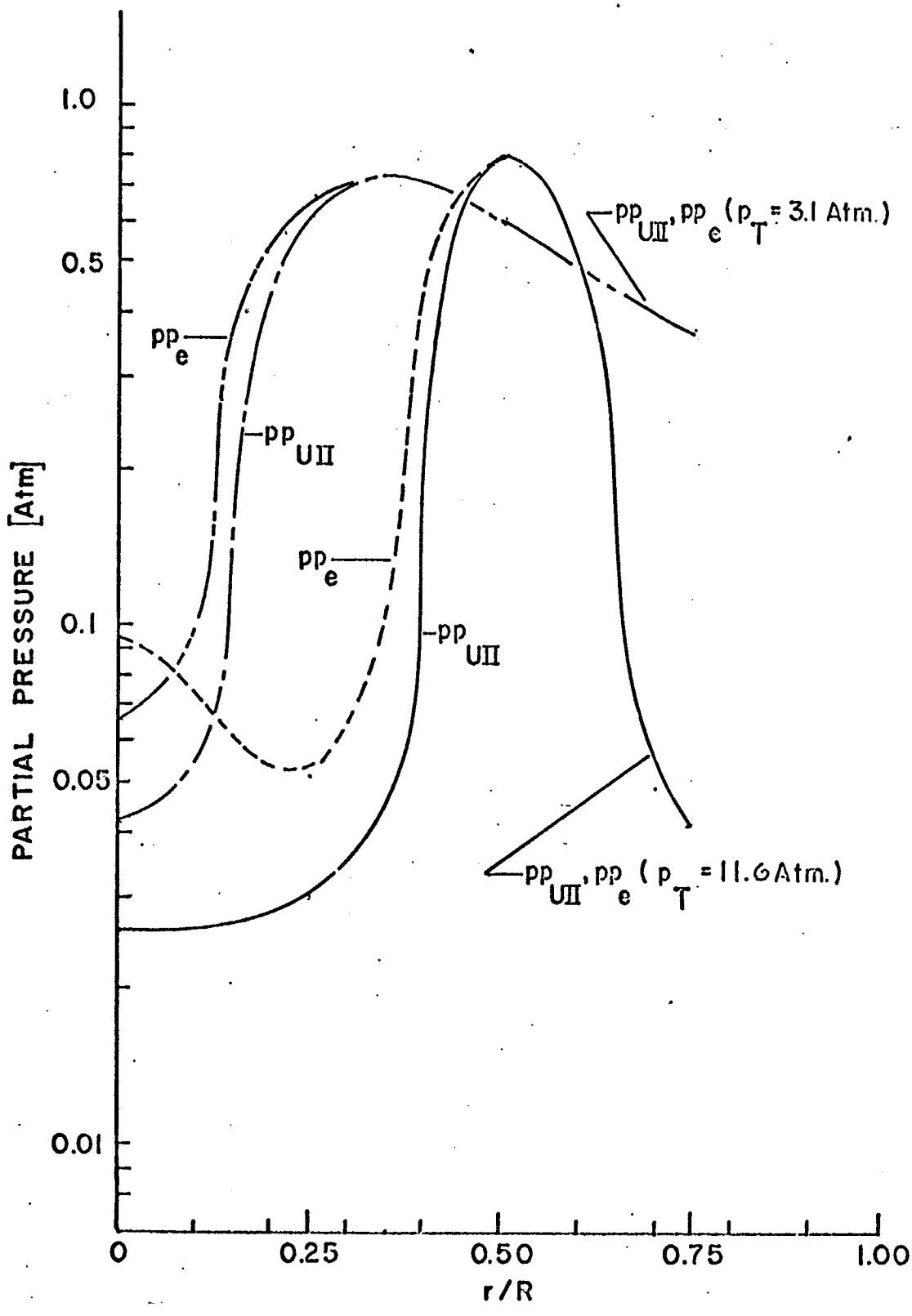

FIG. 8 URANIUM II AND ELECTRON PARTIAL PRESSURE PROFILES

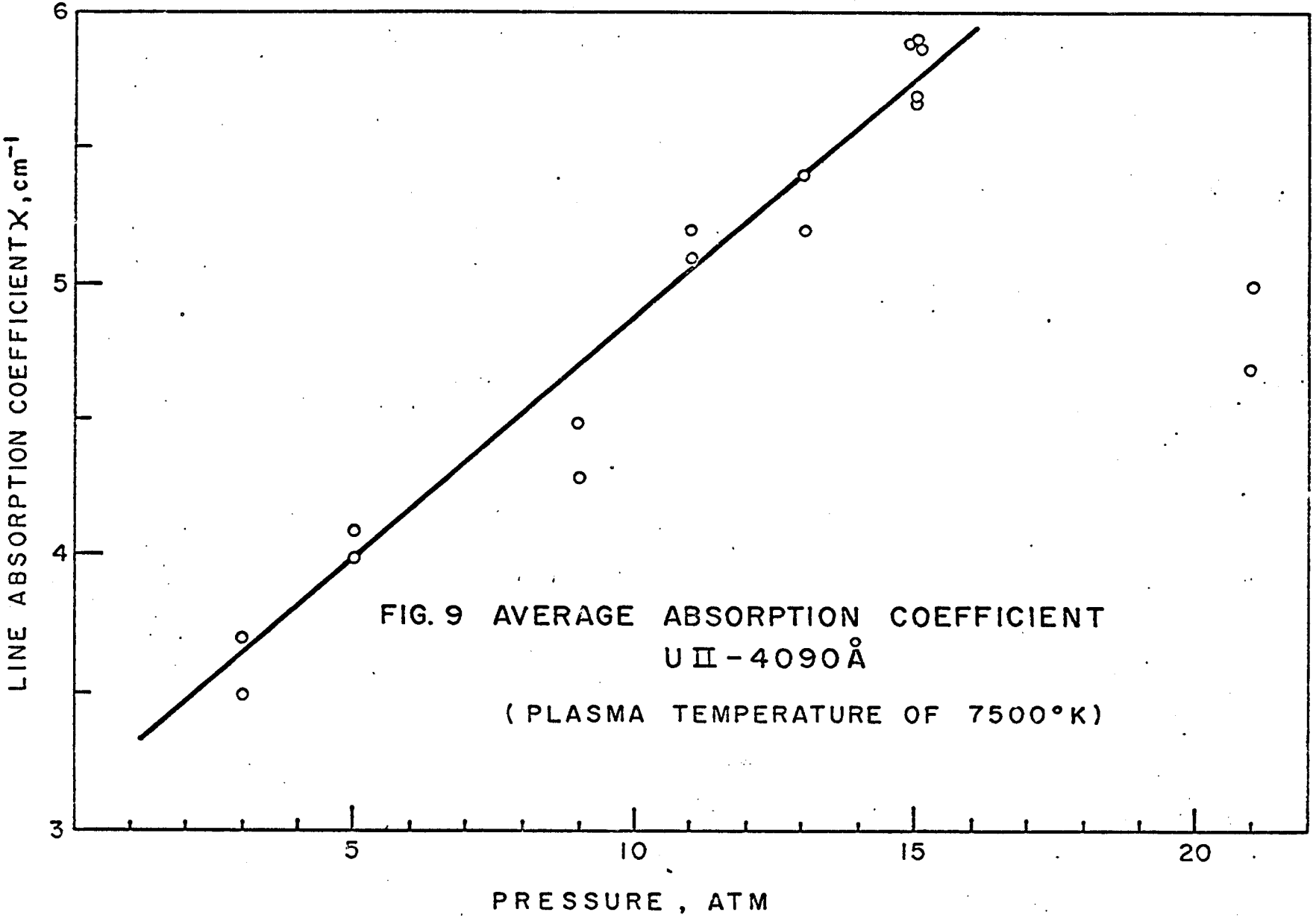

FIG. 9 AVERAGE ABSORPTION COEFFICIENT
U II - 4090 Å
(PLASMA TEMPERATURE OF 7500°K)

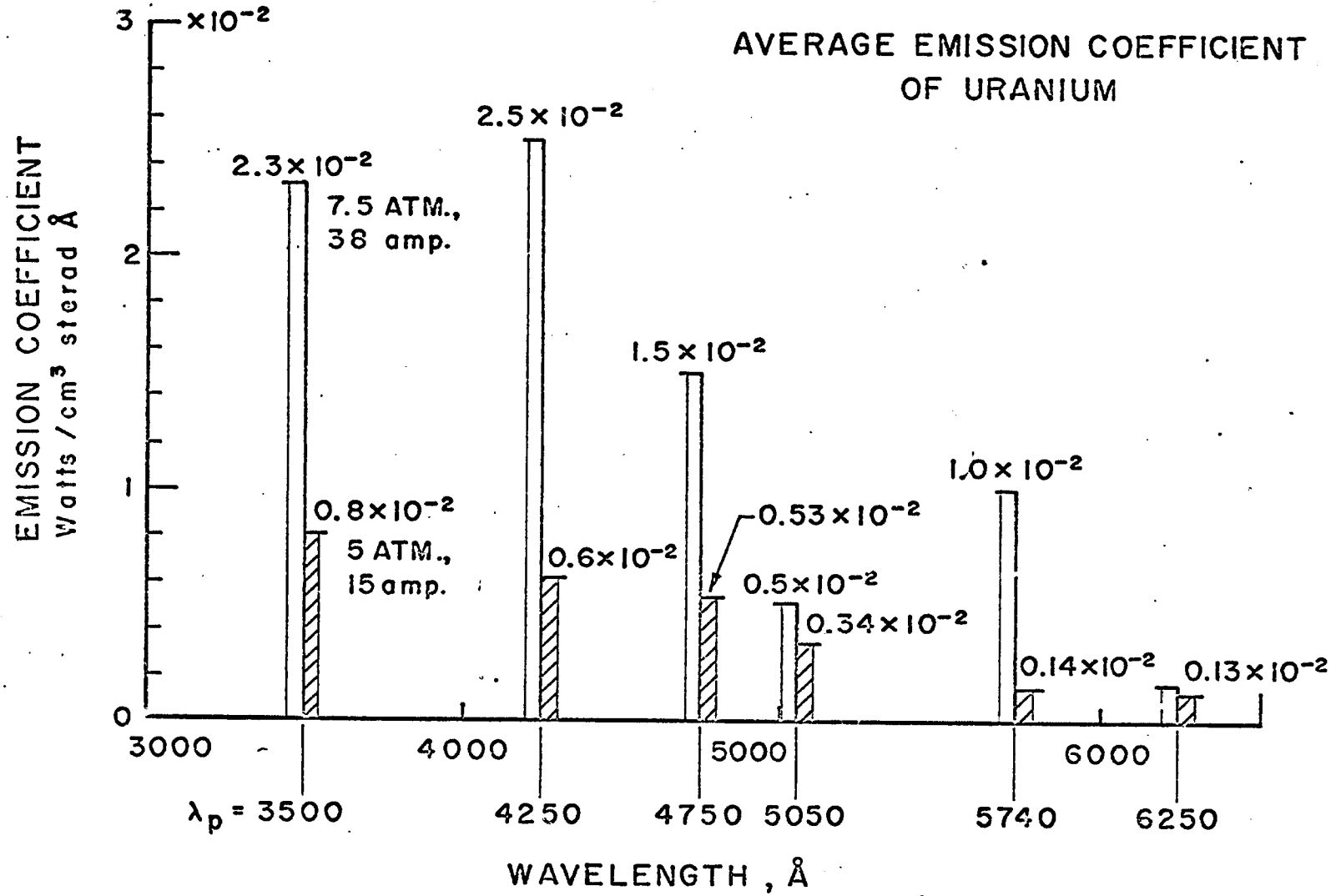
AVERAGE EMISSION COEFFICIENT
OF URANIUM
EMISSION COEFFICIENT
Watts / cm³ sterad Å
3
2
1
0
×10⁻²
2.3×10⁻²
7.5 ATM.,
38 amp.
0.8×10⁻²
5 ATM.,
15 amp.
2.5×10⁻²
0.6×10⁻²
1.5×10⁻²
0.53×10⁻²
0.5×10⁻²
0.34×10⁻²
1.0×10⁻²
0.14×10⁻²
0.13×10⁻²
3000
4000
5000
6000
λp = 3500
4250
4750
5050
5740
6250
WAVELENGTH , Å

FIG. 10

GENERATION OF A FISSIONING PLASMA

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INTRODUCTION

The measurement of the radiatior emission properties of a uranium plasma such as those by LASL (ref. 1), by United Aircraft (ref. 2), and at the University of Florida (ref. 3), have been made on thermally excited plasmas. That is, an electrical discharge, either RF or DC, is used to supply energy to the plasma by electron-atom, electron-ion collisions. These plasmas are essentially high pressure, collision dominated, LTE plasmas.

However, in a plasma core reactor, the fission process is the source of energy. The energy appears in the form of kinetic energy of the two fission fragments or products. The heavy fission fragment has a kinetic energy of approximately 65 Mev, and the light fission fragment has approximately 100 Mev of energy. This is considerably above the thermal energy of the uranium atoms and ions, which is characterized by 1/2 to possibly 3 ev. The process of slowing down and thermalizing these fission fragments will cause extensive nonequilibrium ionization and excitation. Many additional stages of ionization and excitation of the uranium will occur. It is possible that the resultant radiation spectrum may be considerably different from the thermal equilibrium spectrum with no fissions occurring. Even more important, we would like to know what fraction of the fission energy is emitted from the plasma as nonequilibrium radiation relative to the amount of fission energy deposited by "heating up" the plasma, and what is the spectrum of the nonequilibrium radiation.

Thus, it was decided to conduct a fissioning uranium plasma experiment. Also, it was decided that we wanted the ability to generate a "pure" uranium plasma. That is, no cover gas or impurities were to be present. The previously mentioned electrically excited plasmas use an inert cover gas. Since we cannot, from a practical standpoint, compute the radiation spectrum from a fission fragment excited plasma, we did not want any emission lines showing up in the spectrum from material other than uranium since we have no way to separate them. Also, with only uranium present, it is possible that we can deduce better values for the emission constants (statistical weights times transition probabilities) while operating without fissions.

To aid this process, and to start out with the simplest system, the Phase I experiments will not use an uranium arc, but will heat the uranium in an enclosed tungsten oven. Thus the uranium plasma temperatures will be accurately determined by measuring the tungsten oven temperature.

There are, to put it mildly, several problems involved with generating a "pure" uranium plasma. The uranium vapor will condense on cold surfaces, therefore, there cannot be any cold surfaces. The vapor pressure of uranium is low even at the highest solid temperatures, and thus the uranium number density is low, causing the fission rate to be very low relative to that possible in metallic uranium. Later, we will use a DC arc to increase the plasma temperature, but there will be a corresponding decrease in the uranium number density. U-235 is expensive, approximately $20/gram for 93% enriched, thus we want to minimize loss of the uranium. There are safety problems involved working with U-235, since it is a radioactive alpha emitter. There are also radiation hazards associated with the experimental device, since it will be activated by neutron absorption.

EXPERIMENTAL DEVICE

Figure 1 shows a schematic of the fissioning uranium plasma device, for the Phase I experiments. It is a stainless steel vacuum chamber, with two side windows, a vacuum system connection, and windows in the top and bottom flanges. The top flange contains the support rods for the internal components, electrical leads for a tungsten heater, a window, and a deflector system to help keep the uranium which escapes from the oven off of the window. The heart of the internal components is the uranium oven. This is a tungsten can, which contains the uranium plasma. Two slots allow radiation (and unfortunately the uranium plasma) to escape from the oven for spectroscopic measurements, otherwise, it is essentially a black body cavity. A tungsten wire heater surrounds the oven and three rhenium thermal shields surround the heater. This arrangement will permit oven temperatures of 3000°K with an input power to the heater of approximately 5 Kw.

The thermal shields have two sets of sight holes. This permits the wall of the oven to be viewed through the side windows for temperature measurements. A thermal pile and optical system will be mounted on one window so that the oven temperature can be measured when the device is in a neutron field.

For some experiments, a Langmuir probe will be inserted into the oven through the bottom sight tube.

The device will operate in the water shield tank of the University of Florida Training Reactor. The natural convection of the water will cool the chamber.

CALCULATIONS

Let us now look at some quantities that are calculatable for a uranium plasma. Figure 2 shows the vapor pressure curve for uranium, as discussed earlier today by Randol (ref. 4). The total particle number density and the percentage ionization are also shown. These quantities were calculated assuming thermal equilibrium and a saturated uranium vapor or gas. Note that at 3000°K, the reference temperature for the Phase I experiments, the uranium pressure is approaching 0.1 atmospheres, the number density is approximately 0.5×10^{17} particles/cc, and the degree of ionization is approximately 0.07 percent. Thus, we essentially have a neutral uranium gas at 3000°K.

To get an indication of the intensity of the radiation emitted by this thermal equilibrium 3000°K gas, the intensity of the 3839.6Å line was computed as shown in figure 3. This is one of the prominent atom lines. At 3000°K, it has an intensity of 10^{-2}watts/cm^{3}sr. This is quite adequate for spectroscopic measurements.

If we now turn on the neutrons, figure 4 shows the fission rate for saturation conditions. The scale on the right hand side is for a neutron flux of 10^{11}n/cm^{2}sec. This is the approximate flux level available in the water shield tank of the UFTR reactor. For 3000°K, this results in a fission rate of approximately 3×10^6 fission/cc sec. In a high power reactor or in a plasma core reactor, the flux level will probably be as high as 10^{15}n/cm^{2}sec, leading to a 10^4 increase in the fission rate and thus the fission power.

It is interesting to compare the magnitude of the fission power rate in the uranium plasma with the internal energy of the plasma, as an indication of the increased radiation emission from the plasma caused by fissions. Figure 5 shows an approximate ratio of P_f, the fission power in ev/cc sec, to U, the plasma internal energy in ev/cc.

For a neutron flux of 10^{11}n/cm^{2}sec, at 3000°K, the fission events add approximately 1 percent of the internal energy to the plasma every second. Thus, in a hundred seconds, the energy of the plasma would be doubled, if no

energy escaped as radiation. This "enhancement" of the thermal equilibrium radiation by the fission events should be readily detected, particularly since it should appear as non-equilibrium radiation. For higher flux levels, the fission energy released every second can easily exceed the internal energy content of the plasma, even at high temperatures.

Figure 6 shows the planned measurements with the fissioning uranium plasma device. With no neutrons, but with a hot (3000°K) uranium gas, we will identify (assign wavelengths) all of the emitted lines, including any impurities that may be present. The computerized spectrum analysis system that we have developed and reported in the literature (ref. 5) will enable us to do this quickly. Certain prominent lines will be selected and their intensities determined for later reference. With the temperature known accurately, by measuring the uranium oven temperature, the transition probability times the statistical weight for the line can be calculated and checked against the published values.

A Langmuir probe will be inserted into the uranium oven in some experiments, to compare the electron temperature with the oven temperature. The integrated radiation intensity will be measured, as an indication of the energy content of the plasma.

Now, we may have some problems interpreting the data obtained with no neutrons; that is, the thermal equilibrium data. The plasma is in a black body cavity, but with a line of sight through the cavity. Thus we may see no lines, but just a 3000°K black body spectrum. However, the fact that the line of sight goes completely through the cavity probability means that we won't see a continuous black body spectrum, but rather a spectrum of lines whose wavelengths correspond to the uranium lines and whose intensity corresponds to the intensity of the black body spectrum at 3000°K. Regardless of the outcome, a reference spectrum with no neutrons will be available to compare with the spectrum obtained with fissions occurring.

The device will be placed in neutron field and the measurements repeated. Any new lines in the spectrum will be found by processing the data through the computerized spectrum analysis system. Any shift in the distribution will be checked to see if the various lines are enhanced or maybe even depressed. From this, we hope to be able to determine the degree of non-equilibrium present in the plasma, and the trend for extrapolating to other operating conditions. A Langmuir probe will also be used in some

experiments, particularly as a function of time. Neutron flux calibration measurements will be made so that the fission rate can be determined. The integrated radiation intensity will be measured, although not much change in total radiation is expected for our operating conditions as indicated earlier in figure 5.

FINAL REMARKS

Having discussed our purpose in pursuing a fissioning uranium plasma research program, as well as the experimental device and the planned measurements, it is only appropriate to give a status report. We have accomplished the original planning, design, and construction of the experimental device. Some support equipment is still being assembled. We are starting preliminary checkout and calibration tests. Experiments with no fissions using U-238 should start in the near future, barring the appearance of major problems. We will start experiments with a fissioning uranium plasma later this year. We look forward to some illuminating results.

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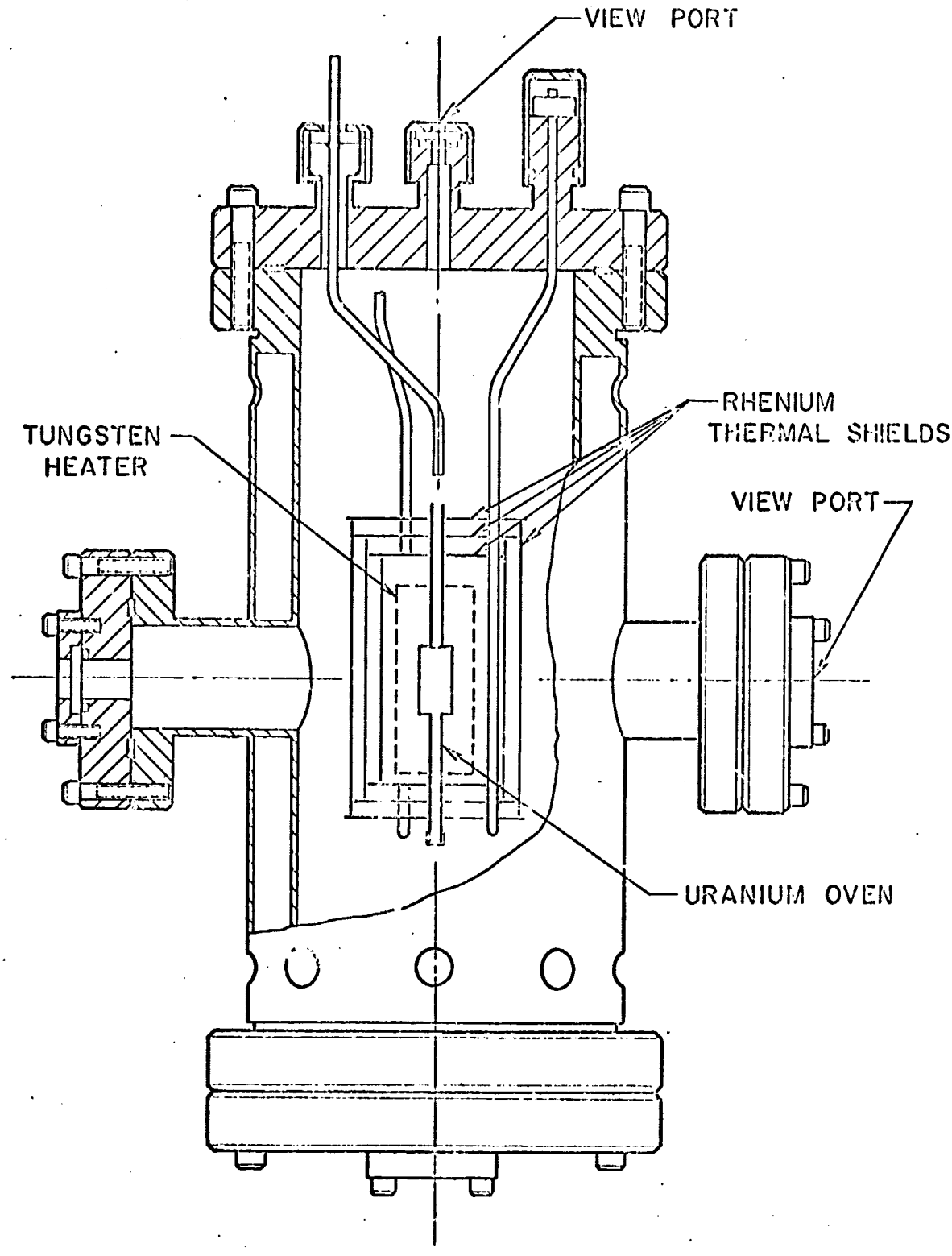

FIG. 1. FISSIONING URANIUM PLASMA DEVICE

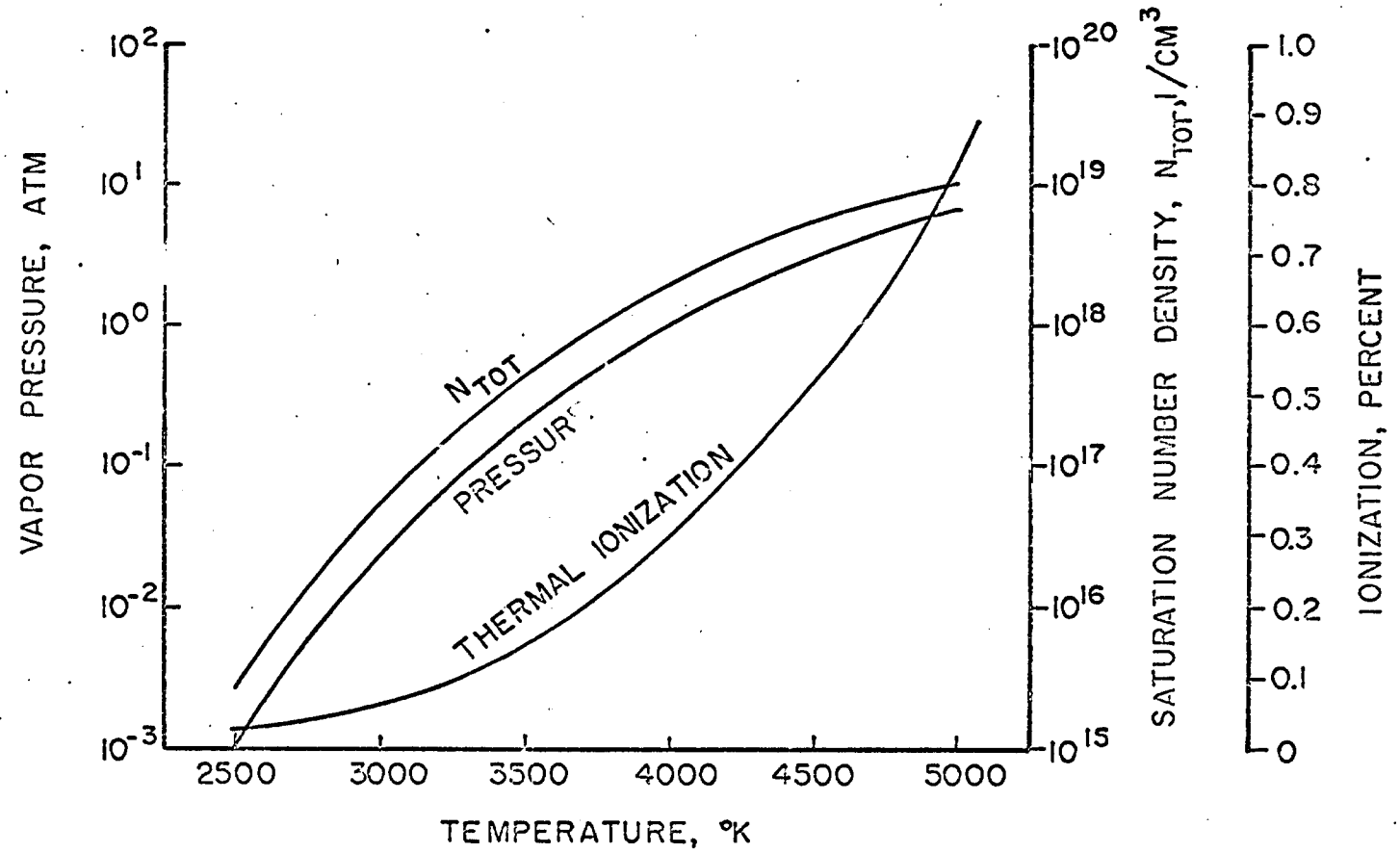

FIG. 2. VAPOR PRESSURE OF URANIUM

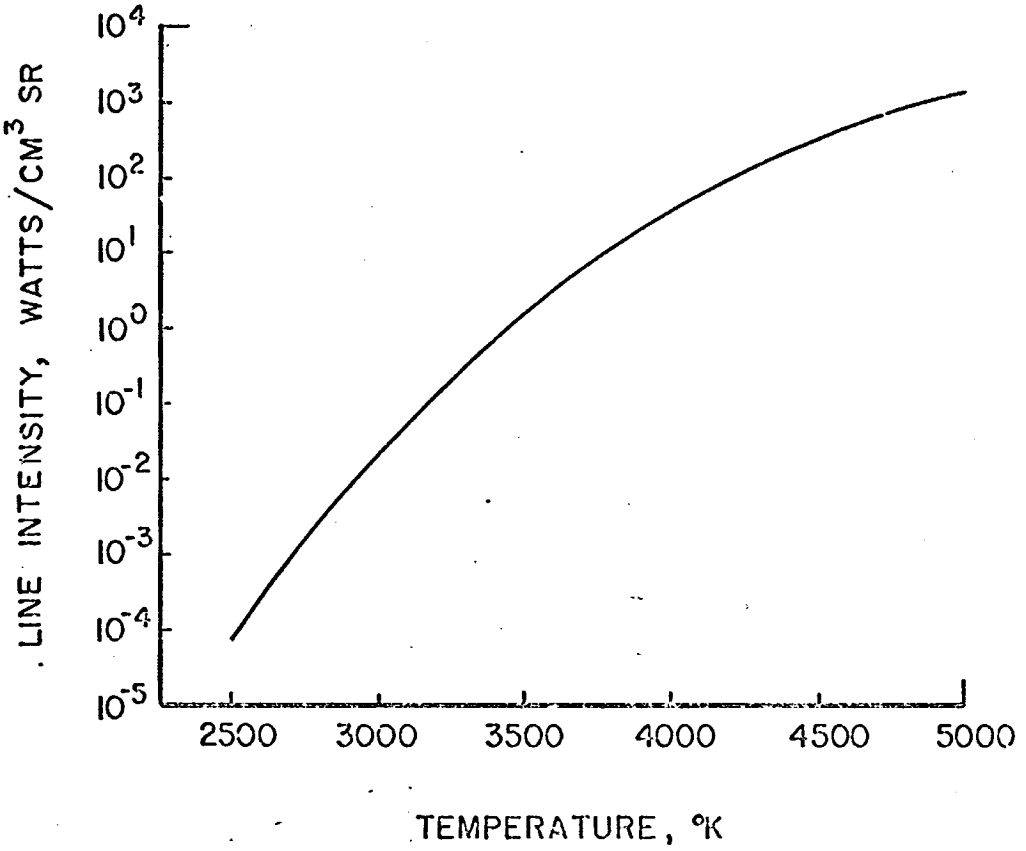

FIG. 3. EMISSION COEFFICIENT OF THE UI 3839.6 Å LINE FOR SATURATION CONDITIONS

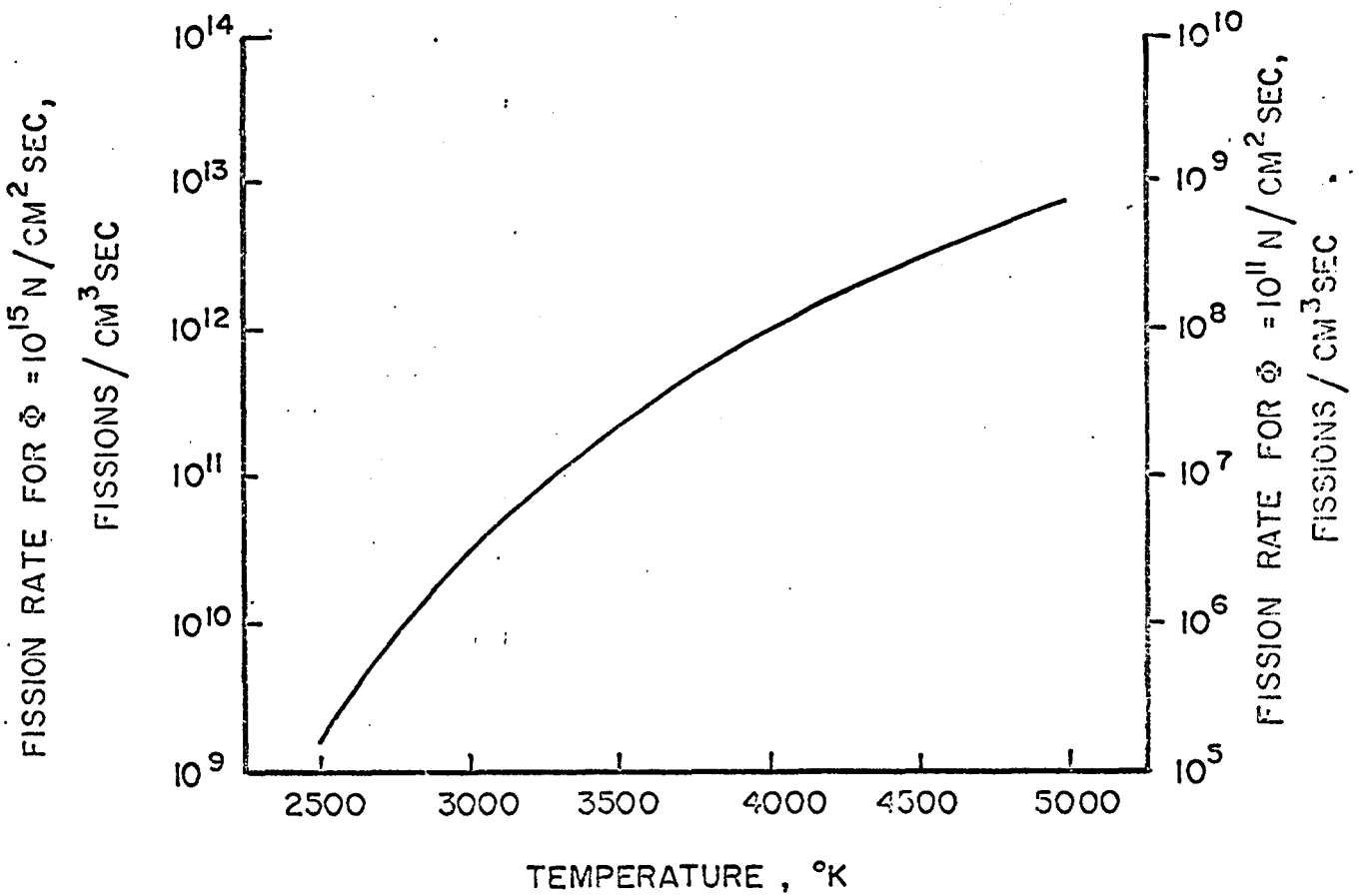

FIG. 4. FISSION RATE FOR U-235 FOR SATURATION CONDITIONS
(THERMAL NEUTRONS 20 °C)

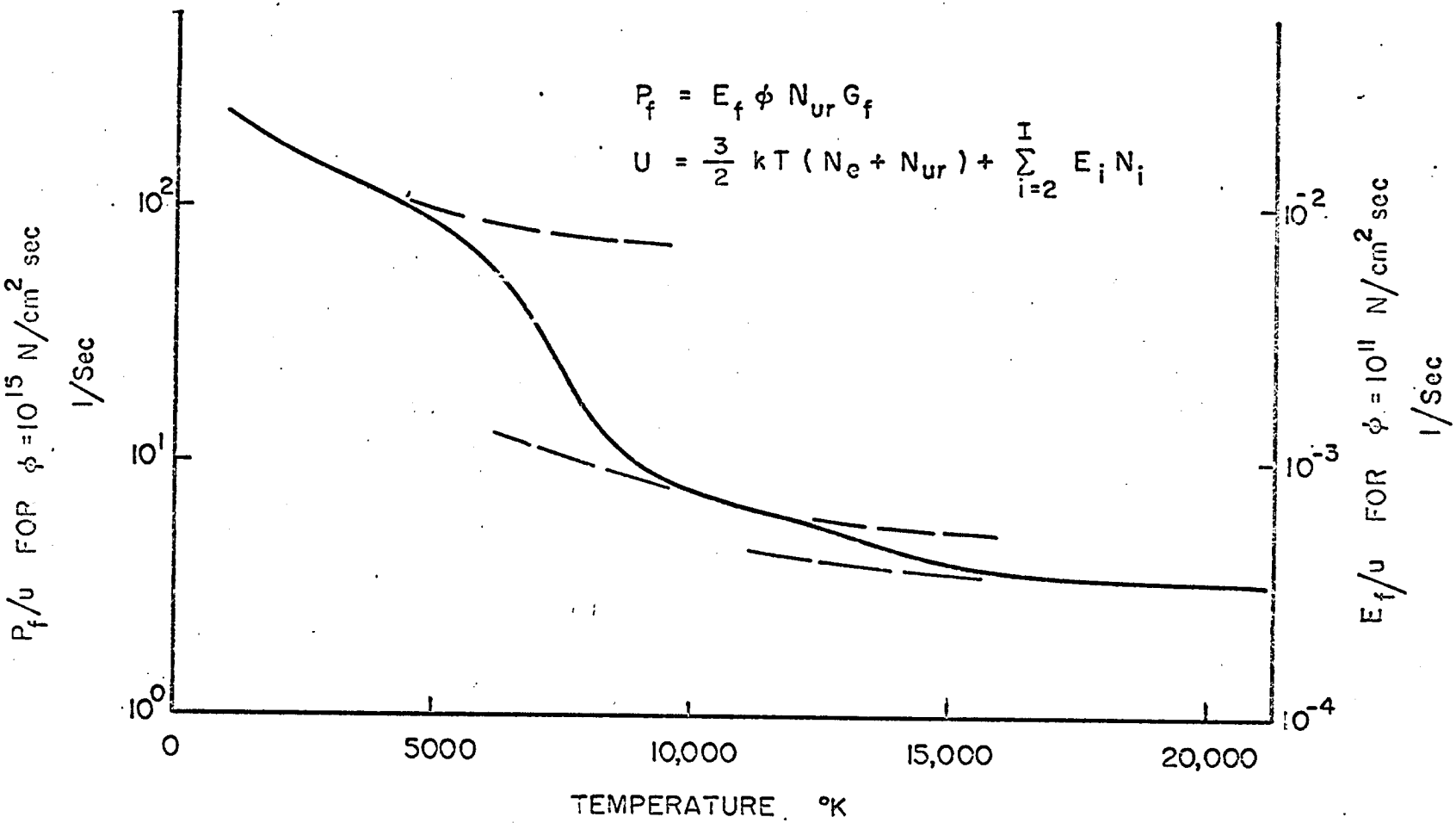

FIG. 5. APPROXIMATE RATIO OF FISSION POWER TO PLASMA ENERGY CONTENT (THERMAL NEUTRONS 20° C)

FIG. 6. MEASUREMENTS

- WITHOUT NEUTRONS
  - IDENTIFICATION OF ALL LINES (IMPURITIES)
  - INTENSITY OF CERTAIN LINES (KNOWN TEMPERATURE)
  - DEDUCE BASIC CONSTANTS
  - LANGMUIR PROBE
  - TOTAL RADIATION
- WITH NEUTRONS
  - IDENTIFY SPECTRUM CHANGE
  - INTENSITY CHANGE OF CERTAIN LINES
  - LANGMUIR PROBE
  - NEUTRON FLUX
  - TOTAL RADIATION