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Electronic Partition Functions for Plasmas in a Capillary Discharge

by John D. Powell

ARL-TN-171 September 2000

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Electronic Partition Functions for Plasmas in a Capillary Discharge

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Abstract

Calculations are undertaken for the electronic partition functions of plasmas in a temperature and pressure range relevant to electrothermal-chemical gun applications. The intent is to assess the limitations of results obtained previously, as well as to provide an improvement for existing models for the capillary discharge. Various thermophysical properties that depend on the electronic partition functions are calculated and compared with results obtained in preceding work. The general conclusion reached is that accurate predictions of the concentrations of minority species in the plasma require highly accurate calculations of the partition functions. Less accurate approximations, however, predict the concentration of majority species, as well as most of the thermophysical properties of the plasma, reasonably accurately.

Acknowledgments

The author is grateful to Dr. William Oberle and Dr. Michael McQuaid for their thoughtful reviews and helpful comments.

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1. Introduction

The purpose of this note is to undertake calculations for the electronic partition functions of plasmas composed of the constituents of polyethylene. Since the calculations are more accurate than those of our previous work, the analysis is a means of evaluating the limitations of existing models and/or extending their applicability to a wider range of temperatures and pressures. Only polyethylene is considered because of its wide use as a plasma-producing liner in capillaries designed for electrothermal-chemical (ETC) launchers. However, it is expected that the general conclusions of this note should apply qualitatively to other substances as well.

In addition to the general desire to provide calculations that are as accurate and internally consistent as possible, undertaking this calculation has also been motivated by some recent work performed by McQuaid and Nusca (1999). In that work, the authors noted that while the approximations employed in our earlier work seemed adequate for the cases of interest then, they might prove inadequate under certain other conditions (the details of which will be discussed subsequently). The intention of the present work is two-fold. The first goal is to indicate what specific plasma properties are not adequately predicted with the approximations in the existing models and under what conditions that failure occurs. The second goal is to perform more accurate calculations and include the methodology for general use in the existing models. We will refer extensively to our previous work (Powell and Zielinski 1992, 1993) in order to avoid needless repetition. The first of these references is referred to as PZ1.

Various electrical and thermodynamic properties of a plasma depend on the electronic partition functions of its constituents. This function for any particular monatomic constituent α can be written as (see equation [24], PZ1)

$$Z_{j\alpha} = \sum_i g_{j\alpha i} \exp(-U_{j\alpha i}/kT), \tag{1}$$

where $U_{j\alpha i}$ is the energy level of the i^{th} electronic state of the j^{th} ion, and $g_{j\alpha i}$ is the appropriate degeneracy factor for this level. The actual calculation in equation (1) is problematic for several reasons. First, for an isolated atom or ion, the sum diverges. This divergence arises because there are an infinite number of discrete states, and the levels $U_{j\alpha i}$ approach a constant value, namely, the ionization energy, as i becomes large. This problem is somewhat illusory since the radii of the electronic orbits increase with increasing energy, eventually becoming comparable to the interatomic spacing. At this point, the atom is essentially ionized. The effect is generally modeled by reducing the ionization potential of the isolated atom or ion by an amount that depends on the density of the mixture, and by including in the summation only the terms whose energy is less than the reduced ionization energy. The sum therefore remains finite. However, general agreement does not exist on the amount by which the ionization potential should be reduced.

The second problem arises because the energy levels in equation (1) are inadequately known either experimentally or theoretically. Most investigators rely on the data contained in Moore's famous tables (Moore 1971), but many of the levels, particularly those that correspond to high orbital angular momentum quantum numbers, have never been observed experimentally and are missing from the tables. Thus, one is forced to either ignore the missing levels (Kovitya 1985) or estimate their values from rather crude theoretical considerations.

Simpler approximations are also common in calculations involving the partition functions. In some cases (Loeb et al. 1987; Mohanti 1990), the ratio of various functions that occurs in certain equations is replaced by unity as a crude approximation, and the sum is not even evaluated. Other authors employ only the first few terms (Cambel 1963; Zeldovich and Raizer 1966), or even just the leading term (i.e., the degeneracy of the ground state) (Zaghloul et al. 2000; Ngo et al. 1998) in the sum in equation (1). Presumably, these approximations that employ only a few terms are made with the belief that the higher energy states are not appreciably populated. While the approximations frequently seem to work, they are difficult to justify in a rigorous sense because the degeneracy factors increase with increasing principal

quantum numbers. Thus, although the higher energy states are not highly populated, they may have a significant effect because of their greater number.

In our earlier calculations of plasma properties for both electromagnetic (Powell and Batteh 1981) and electrothermal (Powell and Zielinski 1992, 1993) launchers, only the first few terms were generally included in the partition-function calculations. For the polyethylene plasma, only those terms from Moore's tables that had energy less than about 9 ev were included. Some studies were undertaken in which the number of terms was varied and only minimal changes in the plasma properties were observed, but these studies were not extensive. Furthermore, McQuaid and Nusca (1999) have pointed out that under some conditions, discrepancies exist between the magnitude of partition functions calculated with only those terms retained and the functions computed by Drawin and Felenbok (1965). The discrepancy occurs at high temperatures and at pressures generally lower than those relevant to our calculations. In the interest of being as complete and thorough as possible, we sought to obtain better approximations for the functions and to determine their impact on various plasma properties.

2. Calculations

As in PZ1, the plasma is assumed to be composed completely of the monatomic constituents: H, H^+, C, C^+, C^{++}, and e^-. For polyethylene, there are twice as many hydrogen heavy particles (ions and neutrals) as carbon heavy particles. As before, we also assume that the ionization potential for the j^{th} ion, $I_{j\alpha}$, is reduced because of the finite density by an amount $\Delta I_{j\alpha}$ (Ebeling and Sandig 1973) given by

$$\Delta I_{j\alpha} = \frac{je^2}{4\pi\varepsilon_0\left(\lambda_D + \overline{\Lambda}/8\right)}. \tag{2}$$

In this equation, e represents the electron charge and ε_0 the permittivity of free space. The parameters λ_D and $\overline{\Lambda}$ are the Debye length and de Broglie wavelength, respectively. These

quantities depend on the temperature and pressure of the plasma; therefore, they must be calculated self-consistently with the other plasma properties. Explicit formulas for their calculation are given in equations (20) and (21) in PZ1. In contrast to previous calculations in which the sum in equation (1) was restricted, included in this calculation are all terms whose energy is less than the reduced ionization potential (i.e., $I_{j\alpha}$- $\Delta I_{j\alpha}$).

For hydrogen, all of the relevant states needed have been calculated, and they are in Moore's (1971) tables. Thus, computation of the function Z_{0H} poses no problem. For carbon, on the other hand, Moore lists levels that have been observed experimentally, and the tables are incomplete. In particular, some levels corresponding to high energy states had not been observed at the time the tables were compiled, and those levels are missing. To rectify that situation, McQuaid and Nusca (1999) attempted to fill in the missing levels both with experimental data from Stiganov and Sventitskii (1968), as well as with their own estimates. Their table, compiled for neutral carbon, is employed in the current calculation to compute the function Z_{0C}. For Z_{1C} and Z_{2C} (i.e., the functions that correspond to first- and second-ionized carbon), only the terms that have been observed and are included in Moore's tables have been accounted for. McQuaid and Nusca (1999) employed a similar approximation, actually including those levels corresponding to an energy less than about 11 ev. They have reasonably argued that the higher energy states should contribute negligibly in the temperature range of interest. However, in the present work the higher levels were included anyway, but the sums were truncated at $I_{1C} - \Delta I_{1C}$ and $I_{2C} - \Delta I_{2C}$. All of the levels in the calculations for the various functions are indicated in the Appendix.

Calculations of all plasma properties have been performed for temperatures ranging from 1 to 3.5 ev and pressures ranging from 1 to 400 MPa. Quantities calculated for each temperature and pressure include: (1) the internal energy E, (2) the density ρ, and (3) the ionization factors x_{1H}, x_{1C}, and x_{2C}. These factors represent the ratio of first or second ions for the particular species to the total number of heavy particles for that species. All of these quantities can be calculated using equations (15)–(23), as well as equation (25), of PZ1. It is necessary, however, to solve

for the various properties iteratively, since the nonlinear, algebraic equations that determine their values are coupled. Only results are presented, and the reader is referred to PZ1, in which a similar, but more complicated, calculation is undertaken for details.

3. Results and Discussion

In Figures 1 and 2, E and ρ are plotted as a function of the temperature and pressure, and in Figures 3–5 are the ionization factors x_{1H}, x_{1C}, and x_{2C}. These particular variables were selected for plotting since most other quantities of interest, such as the electrical conductivity or electron density, follow in a very simple way. These quantities are plotted as a function of temperature T at four different pressures P. The solid curves correspond to results obtained when only levels less than 9 ev were included in the partition functions, as was done in the original calculations; the dotted curves correspond to results obtained when all levels less than the reduced ionization potential were included in the manner described previously.

The results of the calculations exhibit fairly obvious behavior. The internal energy increases with increasing temperature at a fixed pressure, and it decreases with increasing pressure at a fixed temperature. The latter result follows because as the pressure increases, the interatomic spacing becomes smaller, and collisions that tend to deionize the gas become more prevalent. Consequently, at low pressures a substantial part of the energy of the plasma is associated with the energy required to produce ionization. In a similar manner, the ionization factors x_{1H} exhibit the same type of behavior. For carbon, similar behavior is observed at lower temperatures, but eventually second ionization becomes important, and the relative number of first ions is reduced. At the high temperatures and very low pressures considered, particularly at P=1 MPa with T greater than about 2.5 ev, third ionization is probably important, but it is not accounted for in the model. However, such low pressures are generally outside the range of interest for electrothermal or electromagnetic gun applications. The mass density decreases with increasing temperature at constant pressure to satisfy the equation of state.

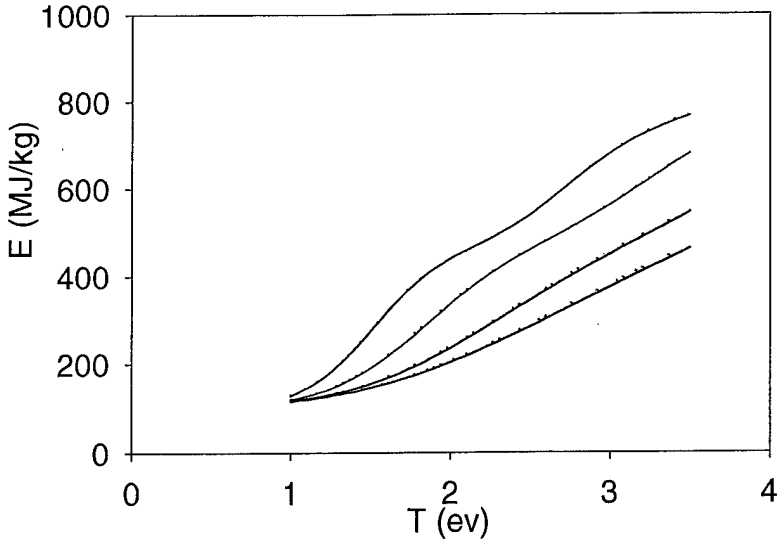

Figure 1. **Internal Energy as a Function of Temperature and Pressure. Solid curves represent results when only levels less than 9 ev are retained in the calculations; dotted curves represent results when all terms less than the reduced ionization potential are retained. Pressures are denoted by: black, P = 1 MPa; red, P = 10 MPa; blue, P = 100 MPa; and green, P = 400 MPa.**

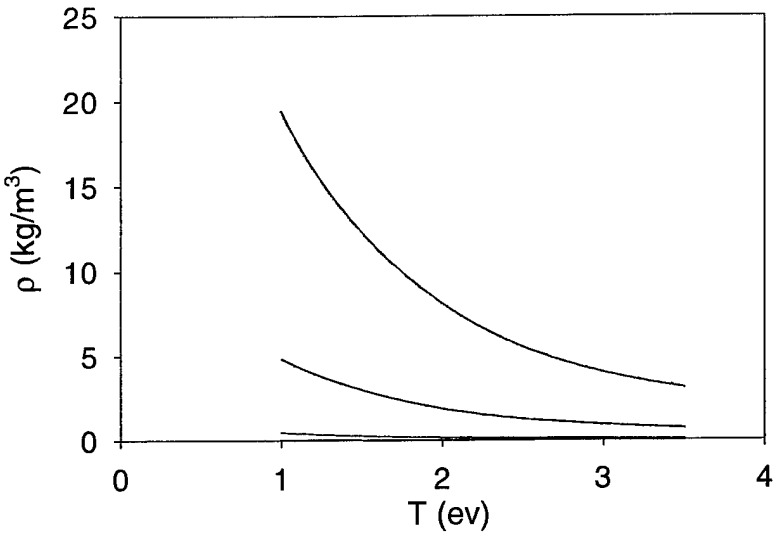

Figure 2. **Density as a Function of Temperature and Pressure. Solid curves represent results when only levels less than 9 ev are retained in the calculations; dotted curves represent results when all terms less than the reduced ionization potential are retained. Pressures are denoted by: black, P = 1 MPa; red, P = 10 MPa; blue, P = 100 MPa; and green, P = 400 MPa.**

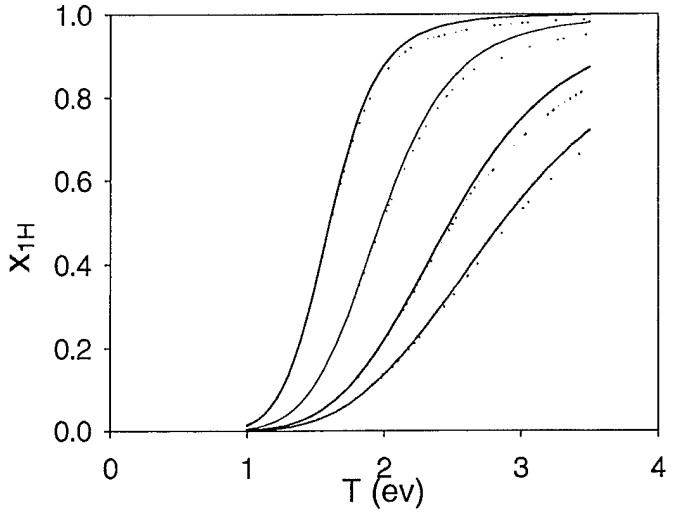

Figure 3. First Ion Concentration of Hydrogen as a Function of Temperature and Pressure. Solid curves represent results when only levels less than 9 ev are retained in the calculations; dotted curves represent results when all terms less than the reduced ionization potential are retained. Pressures are denoted by: black, P = 1 MPa; red, P = 10 MPa; blue, P = 100 MPa; and green, P = 400 MPa.

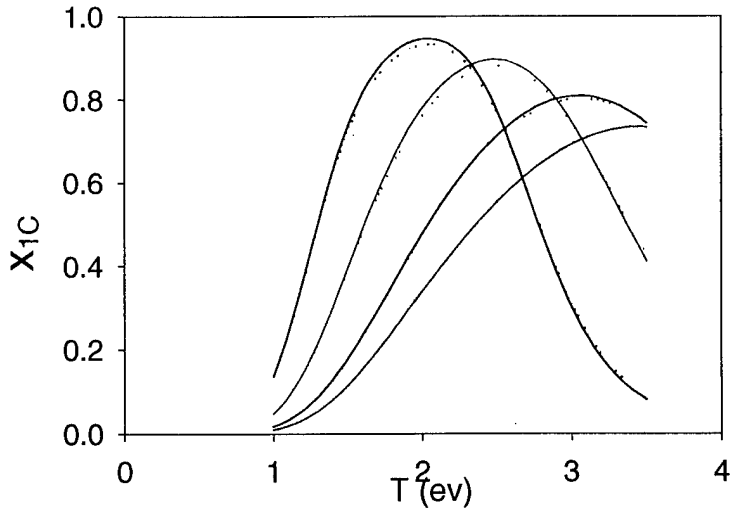

Figure 4. First Ion Concentration of Carbon as a Function of Temperature and Pressure. Solid curves represent results when only levels less than 9 ev are retained in the calculations; dotted curves represent results when all terms less than the reduced ionization potential are retained. Pressures are denoted by: black, P = 1 MPa; red, P = 10 MPa; blue, P = 100 MPa; and green, P = 400 MPa.

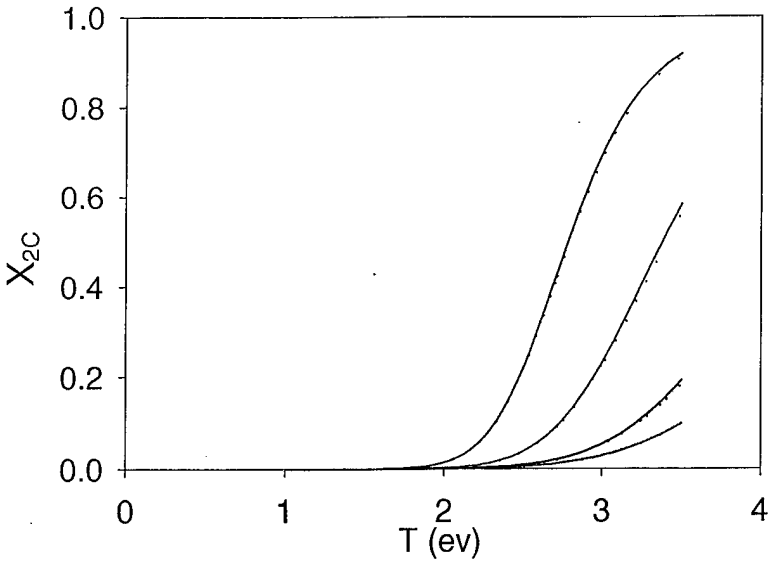

Figure 5. Second Ion Concentration of Carbon as a Function of Temperature and Pressure. Solid curves represent results when only levels less than 9 ev are retained in the calculations; solid curves represent results when all terms less than the reduced ionization potential are retained. Pressures are denoted by: black, P = 1 MPa; red, P = 10 MPa; blue, P = 100 MPa; and green, P = 400 MPa.

Of the greatest interest in the results is the very weak dependence on the number of terms included in the partition functions. In most cases, differences are imperceptible. For example, consider the results for x_{1H} at P = 100 MPa and T = 3 ev, a point where one of the greater deviations is observed. Superscript s (for solid) denotes results for the original calculation, and superscript d (for dotted) denotes results for the case in which all terms are included in the partition functions. At the temperature and pressure in question we find $x^s_{1H} = 0.7555$ and $x^d_{1H} = 0.7076$, a difference of only about 7%. On the other hand, the partition functions at this point (not plotted), given by $Z^s_{0H} = 2.000$ and $Z^d_{0H} = 2.604$, differ by 23%. The reason that significant differences in the partition functions for the two calculations lead to only small differences in the relative number of ions can be described as follows. At temperatures sufficiently high that there is appreciable population of the higher-energy states (i.e., the terms omitted in the calculation of Z^s_{0H} should not really have been neglected), most of the hydrogen atoms (71%) have already been ionized. Thus, if the temperature is increased still more so that

Z^d_{0H} is even larger relative to Z^s_{0H}, minimal changes will be observed in x^d_{1H} since its value can never exceed unity. To make the argument more concrete, consider the values obtained at the same pressure and at $T = 3.5$ ev, where $x^s_{1H} = 0.8720$, $Z^s_{0H} = 2.000$, $x^d_{1H} = 0.8222$, and $Z^d_{0H} = 2.990$. For this case, the percentage differences in the partition functions are 33%, whereas the differences in the ionization factors are only 6%. Similar conclusions apparently hold (perhaps even more so) for carbon, except that x_1 approaches zero rather than unity at high temperatures when the second ionization begins to dominate.

It is important to note that while these conclusions hold for the majority species, they do not hold for the minority species. Thus, the concentrations of neutral hydrogen predicted at $T = 3$ ev are $x^s_{0H} = 0.2445$ and $x^d_{0H} = 0.2924$; at 3.5 ev the corresponding values are $x^s_{0H} = 0.1280$ and $x^d_{0H} = 0.1778$. Here, the percentage differences are 16% and 28%, respectively, and the situation gets worse as the temperature increases.

Rolader (1987) reached similar conclusions in a careful study of partition function calculations for aluminum and copper, two substances that are important in plasma armatures for electromagnetic guns. In particular, he considered three different methods of performing the calculations. In the first, only levels given in Moore's tables were considered; in the second and third, two different approximations (hydrogenic and Rydberg-Ritz) were employed to fill in the missing levels. In all cases, the partition function series were terminated at the reduced ionization potentials. At high temperatures, where the higher-energy states were most populated, and at low pressures, where the reduction in ionization potential was small, the number and magnitude of the terms included in the sum varied greatly. The final conclusion, however, was that in any event, only the minority species were significantly affected. Fortunately, most of the electrical and thermodynamic properties of the plasma do not depend sensitively on the minority species. It is for this reason that we can be somewhat cavalier in calculating these functions. Rolader, however, pointed out that minority species can be important in calculating the opacity of the plasma, and that spectroscopic studies could focus on minority species whose probabilities

may be better known than those of the majority species. For studies such as these, detailed and careful calculations of the partition function would clearly be required.

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Appendix:

Energy-Level Data Employed in Calculations

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Table A-1 contains the data employed in calculations of the electronic partition functions for H, C, C^+, and C^{++}. These data were obtained from the sources indicated earlier. In some cases, several levels of comparable energy are combined into a single level denoted by i with degeneracy g. Maximum values included in the tables are always greater than the reduced ionization potential for any of the calculations undertaken.

Table A-1. Energy-Level Data Employed in Partition-Function Calculations for Hydrogen and Carbon

H j = 0 I_{1H} = 13.60 ev			C j = 0 I_{1H} = 11.26 ev								
i	g	Energy (ev)	i	g	Energy (ev)	i	g	Energy (ev)	i	g	Energy (ev)
1	2	0.00	1	1	0.00	29	5	10.14	57	21	10.88
2	8	10.20	2	3	0.000203	30	1	10.20	58	15	10.88
3	18	12.09	3	5	0.000539	31	5	10.35	59	3	10.89
4	32	12.75	4	5	1.26	32	9	10.38	60	84	10.89
5	50	13.06	5	1	2.68	33	21	10.39	61	9	10.89
6	72	13.22	6	5	4.18	34	15	10.40	62	7	10.89
7	98	13.32	7	9	7.48	35	3	10.40	63	9	10.89
8	128	13.39	8	3	7.69	36	7	10.41	64	3	10.89
9	162	13.43	9	15	7.95	37	3	10.42	65	108	10.89
10	200	13.46	10	3	8.54	38	84	10.42	66	132	10.90
11	242	13.49	11	15	8.64	39	9	10.43	67	36	10.94
12	288	13.51	12	3	8.77	40	3	10.52	68	21	10.98
—	—	—	13	9	8.85	41	15	10.54	69	15	10.99
—	—	—	14	5	9.00	42	12	10.54	70	7	10.99
—	—	—	15	1	9.17	43	5	10.59	71	3	10.99
—	—	—	16	9	9.33	44	1	10.62	72	9	10.99
—	—	—	17	5	9.63	45	5	10.69	73	12	10.99
—	—	—	18	9	9.68	46	21	10.70	74	5	10.99
—	—	—	19	21	9.70	47	9	10.70	75	36	11.02
—	—	—	20	15	9.71	48	15	10.71	76	480	11.04
—	—	—	21	3	9.71	49	3	10.72	77	720	11.05
—	—	—	22	7	9.74	50	7	10.72	78	12	11.05
—	—	—	23	3	9.76	51	84	10.72	79	36	11.07
—	—	—	24	9	9.84	52	3	10.72	80	924	11.10
—	—	—	25	15	9.94	53	9	10.72	81	12	11.12
—	—	—	26	3	9.99	54	108	10.74	82	36	11.14
—	—	—	27	3	10.06	55	36	10.79	83	1152	11.14
—	—	—	28	9	10.08	56	5	10.87	—	—	—

Table A-1. Energy-Level Data Employed in Partition-Function Calculations for Hydrogen and Carbon (continued)

C^+ j = 1 I_{2C} = 24.38 ev						C^{++} j = 2 I_{3C} = 47.86 ev					
i	g	Energy (ev)	i	g	Energy (ev)	i	g	Energy (ev)	i	g	Energy (ev)
1	2	0.00	21	6	22.13	1	1	0.00	34	3	42.57
2	4	0.00793	22	14	22.19	2	9	6.49	35	5	42.68
3	12	5.34	23	4	22.48	3	3	12.69	36	1	42.79
4	10	9.29	24	12	22.53	4	9	17.04	37	7	42.84
5	2	11.97	25	10	22.57	5	5	18.09	38	20	42.97
6	6	13.72	26	2	22.82	6	1	22.63	39	9	42.98
7	2	14.45	27	4	22.86	7	3	29.53	40	5	42.99
8	6	16.33	28	6	22.90	8	1	30.65	41	3	42.99
9	4	17.61	29	8	23.12	9	3	32.11	42	21	43.04
10	10	18.05	30	10	23.39	10	9	32.20	43	7	43.26
11	10	18.66	31	2	24.13	11	15	33.48	44	3	43.99
12	2	19.50	32	4	24.27	12	5	34.28	45	3	44.28
13	6	20.15	33	6	24.27	13	9	38.22	46	7	44.40
14	12	20.71	34	8	24.28	14	3	38.37	47	20	44.47
15	10	20.85	35	10	24.28	15	3	38.44	48	9	44.48
16	6	20.92	36	2	24.37	16	1	38.65	49	5	44.48
17	14	20.95	37	4	24.37	17	9	39.40	50	9	44.49
18	2	21.50	38	6	24.37	18	3	39.65	51	7	44.53
19	20	21.74	39	8	24.37	19	15	39.85	52	3	45.08
20	6	22.10	—	—	—	20	21	39.92	53	3	45.25
—	—	—	—	—	—	21	3	39.98	54	15	45.33
—	—	—	—	—	—	22	7	40.01	55	5	45.39
—	—	—	—	—	—	23	15	40.06	56	3	45.87
—	—	—	—	—	—	24	5	40.20	57	15	45.93
—	—	—	—	—	—	25	3	40.58	58	15	46.34
—	—	—	—	—	—	26	9	40.88	59	5	46.70
—	—	—	—	—	—	27	5	41.25	60	3	47.26
—	—	—	—	—	—	28	5	41.31	61	5	47.36
—	—	—	—	—	—	29	21	41.34	62	7	47.36
—	—	—	—	—	—	30	15	41.86	63	3	47.65
—	—	—	—	—	—	31	3	42.15	64	5	47.66
—	—	—	—	—	—	32	9	42.17	65	5	47.82
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Calculations are undertaken for the electronic partition functions of plasmas in a temperature and pressure range relevant to electrothermal-chemical gun applications. The intent is to assess the limitations of results obtained previously, as well as to provide an improvement for existing models for the capillary discharge. Various thermophysical properties that depend on the electronic partition functions are calculated and compared with results obtained in preceding work. The general conclusion reached is that accurate predictions of the concentrations of minority species in the plasma require highly accurate calculations of the partition functions. Less accurate approximations, however, predict the concentration of majority species, as well as most of the thermophysical properties of the plasma, reasonably accurately.

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