STEADY-STATE COMPOSITION OF A LOW-DENSITY NONEQUILIBRIUM HYDROGEN PLASMA

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SUMMARY

The steady-state composition of a hydrogen plasma is computed for both Maxwellian and non-Maxwellian electron energy distributions for the case in which only volume effects need be considered. The electron energy distribution function is so defined that it can be smoothly varied from Maxwellian to Druyvesteyn. Steady-state plasma equations are derived and are solved numerically for selected forms of this energy distribution. The results apply for plasmas that have a density of less than $10^{13}$ particles per cubic centimeter. The gas is shown to be fully ionized for all distribution functions considered when the kinetic temperature of the electrons is above 4 electron volts. Certain regions of electron temperature are shown to exist in which Maxwellian and non-Maxwellian distributions predict substantially different plasma compositions.

INTRODUCTION

The current interest in the production and properties of high-temperature plasmas has prompted renewed studies of the relation between temperature, energy distribution, and composition of such plasmas. One of the earlier attempts was made by Saha (refs. 1 and 2), who formulated a relation between the temperature and the degree of ionization of a three-component (neutral atoms, ions, and electrons) plasma. Saha assumed the three components to behave as ideal monatomic gases and assumed complete thermodynamic equilibrium between the particles and the radiation field. An extension of Saha's work that allowed the radiation to escape from the plasma is described in reference 2.

The analyses mentioned so far are applicable only to monatomic gases. A further treatment, with thermodynamic equilibrium assumed, was made by Gross and Eisen (ref. 3). This treatment considers a plasma composed of hydrogen atoms, molecules,
atomic ions, negative ions, and electrons. The existence of molecular ions is not taken into account.

Dewan (ref. 2) treated the case of a steady-state, nonequilibrium, three-component plasma and showed that the form of the electron energy distribution plays an important role in the relation between the electron temperature and the plasma composition.

This report extends the work of Dewan to the case of a plasma composed of hydrogen molecules, atoms, molecular ions, atomic ions, and electrons. The method used is similar to that of Drawin (ref. 4). Both the effects of electron temperature and electron distribution are considered. The electron energy distribution function is defined in such a manner that it can be smoothly varied from Maxwellian to Druyvesteyn.

The model used imposes certain restrictions on the applicability of the results. The first assumption is that the plasma composition is determined by processes within the plasma volume and that wall recombination is negligible. Since this assumption is not valid for many laboratory plasmas, the results of the present approach would not apply to those plasmas.

A further restriction in the model used herein is that excited states are neglected. This restriction is valid if the probability of a collision of an electron with an excited molecule or atom is small compared with the probability of a collision with a ground-state molecule or atom, which is the case if the plasma density is not too high or if the plasma is optically thin.

The calculations are performed for molecular hydrogen densities of $10^9$ and $10^{13}$ particles per cubic centimeter and kinetic temperatures from 0.4 to 6.0 electron volts. Experimental values have been used for the cross sections and collision frequencies whenever possible.

**SYMBOLS**

- **A** rate coefficient, defined in appendix
- **a** normalizing factor, defined in eq. (15)
- **B** rate coefficient, defined in appendix
- **b** defined in eq. (15)
- **C, C_1, C_2** rate coefficients, defined in appendix
- **D** rate coefficient, defined in appendix
- **E** rate coefficient, defined in appendix
- **F** rate coefficient, defined in appendix
- **F_1** reaction rate of process (11)

2
\( F_2 \) reaction rate for process (f2)

\( k \) Boltzmann constant, ergs/\(^{0}\)K

\( m \) electron mass, g

\( n \) number of particles per cm\(^3\)

\( n_m \) original number of particles per cm\(^3\)

\( T \) temperature, \(^{0}\)K

\( t \) time, sec

\( v \) electron velocity, cm/sec

\( \alpha \) recombination coefficient

\( \langle \alpha_1 \rangle \) radiative recombination coefficient

\( \langle \alpha_2 \rangle \) collisional recombination coefficient

\( \beta \) \( 3.5 \times 10^{16} / \text{(torr)(cm}^3) \)

\( \epsilon \) electron energy, ergs

\( \langle \epsilon \rangle \) average energy, ergs

\( \sigma \) cross section, cm\(^2\)

\( \tau \) kinetic temperature, defined in eq. (16)

\( \nu \) collision frequency, (torr\(^{-1}\))(sec\(^{-1}\))

\( \bar{\nu} \) \( \nu n_{mo} / \beta \)

Subscripts:

\( ac \) atomic collisional

\( ai \) atomic ion

\( ao \) neutral atom

\( ar \) atomic radiative

\( dr \) dissociative recombination

\( e \) electron

\( id \) molecular ion dissociation

\( mi \) molecular ion

\( mo \) neutral molecule

\( rc \) radiative collisional
THEORY

Model

As previously mentioned, the plasma is considered to be optically thin and sufficiently large or well contained so that wall effects are negligible.

Only the following reactions are considered:
(a) \( H + e \rightarrow H^+ + 2e \) (Atomic ionization)
(b) \( H_2 + e \rightarrow H_2^+ + 2e \) (Molecular ionization)
(c1) \( e + H^+ \rightarrow H \) (Radiative recombination)
(c2) \( e + e + H^+ \rightarrow H + e \) (Collisional recombination)
(d) \( e + H_2^+ \rightarrow H + H \) (Dissociative recombination)
(e) \( H_2^+ + e \rightarrow H^+ + H + e \) (Dissociation of molecular ions)
(f1) \( H + H + H_2 \rightarrow H_2^+ + H_2 \) (Three-body atomic recombination)
(f2) \( H + H + H \rightarrow H_2 + H \)

A search of the literature indicated that all other reactions were negligible compared with those included with the possible exception of ionization of excited atoms or molecules. The neglect of this type of ionizing collision can be justified if the density to which the model is applied is properly restricted. Since the decay time of an excited state is of the order of \( 10^{-8} \) second (ref. 5, p. 9), it is sufficient to impose the condition that the time between ionizing collisions be large compared with \( 10^{-8} \) second. With the use of the maximum value of the ionization frequency for the hydrogen molecule, the model is suitable for densities less than about \( 10^{13} \) particles per cubic centimeter. For these densities, the time between ionizing collisions is at least \( 10^{-6} \) second.

At the lower temperature and higher density end of the region treated herein, the plasma may tend to become optically thick for the resonance lines. Thus, the assumption of optical thinness must be carefully checked by independent methods. Other assumptions are that the plasma is considered to be in a steady state, that the ions and the neutrals are at room temperature (charge exchange reactions can be neglected), and that the original gas is composed of diatomic molecules.

PRODUCTION RATES

The number of atomic ions formed per unit time per unit volume by process (a) is

\[
\frac{d}{dt} (n_{ai}) = n_{ao} \int_0^\infty \sigma_{ai} v \, dn_e = an_e n_{ao} A
\]
where \( a \) is a normalizing constant that depends on the distribution function, and \( A \) is the rate coefficient for process (a). (All rate coefficients are evaluated in the appendix.)

Similarly, the number of molecular ions formed per unit time per unit volume by process (b) is

\[
\frac{d}{dt} (n_{mi}) = \int_0^\infty \nu_{mo} \, dn_e = \frac{n_{mo}}{\beta} \int_0^\infty \nu_{mo} \, dn_e = an_e n_{mo} B \quad (2)
\]

The loss rate of atomic ions due to radiative recombination (process (c1)) is

\[
\frac{d}{dt} (n_{ar}) = \langle \alpha_1 \rangle \, n_{ai} n_e = an_e n_{ai} C_1 \quad (3a)
\]

An integration is not performed for this case since only the total recombination coefficient was known. The cross section for this process, however, is expected to approach infinity as the energy approaches zero and is a monotonically decreasing function of energy. Process (d) also exhibits this behavior, and the appendix shows that the form of the distribution function has little effect on the result. Thus, using a rate coefficient that is independent of the energy distribution is not believed to introduce a large error.

A similar consideration holds for collisional recombination (process (c2)), which is written

\[
\frac{d}{dt} (n_{ac}) = \langle \alpha_2 \rangle \, n_{ai} n_e^2 = an_e n_{ai} C_2 \quad (3b)
\]

The rate of atom production due to dissociative recombination (process (d)) is

\[
\frac{d}{dt} (n_{dr}) = 2n_{mi} \int_0^\infty \sigma_{dr} \, v \, dn_e = 2a_{ni} n_{mi} \quad (4)
\]

The rate of production of atoms (or atomic ions) by dissociation of molecular ions (process (e)) is

\[
\frac{d}{dt} (n_{id}) = n_{mi} \int_0^\infty \sigma_{id} \, v \, dn_e = an_e n_{mi} \quad (5)
\]

The rate of loss of atoms due to processes (f1) and (f2) is
\[
\frac{d}{dt}(n_{a0}) = 2n_{a0}^2 n_{mo} F_1 + 2n_{a0}^3 F_2 = 2n_{a0}^2 F
\]  

Here, \( F_1 \) and \( F_2 \) are the reaction rates for processes (f1) and (f2), respectively.

**General Steady-State Equations**

At steady-state conditions, the rate of atomic ion production (processes (a) and (e)) is equal to the rate of atomic ion consumption (processes (c) and (c2)). Thus,

\[
n_{a0} A + n_{mi} E = n_{ai} C
\]  

where

\[
C = C_1 + C_2 n_e
\]

The reactions that give rise to an increase of electrons (processes (a) and (b)) must be balanced by the reactions in which an electron is captured (processes (c) and (d)). Thus,

\[
n_{a0} A + n_{mo} B = n_{ai} C + n_{mi} D
\]

The conservation of neutral atoms requires that

\[
n_{a0} A + \frac{2n_{a0}^2 F}{an_e} = n_{ai} C + n_{mi} (2D + E)
\]  

The only other equations that are needed are those specifying conservation of charge and conservation of mass:

\[
ne = n_{ai} + n_{mi}
\]  

\[
n_m = n_{mo} + n_{mi} + \frac{1}{2} (n_{a0} + n_{ai})
\]

Combining equations (7) and (8) results in
Combining equations (7) and (9) gives

\[
\frac{n_{mi}}{n_{mo}} = \frac{B}{D + E}
\]  

(12)

Combining equations (7) and (9) gives

\[
\frac{n_{ao}^2 F}{a_n e} = n_{mi}(D + E)
\]  

(13)

Equations (7), (10), (11), (12), and (13) are the general steady-state equations that must be solved simultaneously for the amounts of the various components.

Energy Distribution Function

In order to evaluate the coefficients needed to solve the general steady-state equations, the following expression for the electron energy distribution was arbitrarily chosen:

\[
dn_e = a_n e^{-\left(\epsilon/\tau\right)^b} \epsilon^{1/2} \, d\epsilon
\]  

(14)

where \( 1 \leq b \leq 2 \), and \( a \) is a normalizing factor given by

\[
a = \frac{1}{\int_0^{\infty} e^{-\left(\epsilon/\tau\right)^b} \epsilon^{1/2} \, d\epsilon} = \frac{b}{\tau^{3/2} \Gamma\left(\frac{3}{2b}\right)}
\]  

(15)

The factor \( b \) allows the energy distribution to be varied from Maxwellian (\( b = 1 \)) to Druyvesteyn (\( b = 2 \)). The electron kinetic temperature \( \tau \) is related to the electron temperature \( T \) in the following manner:

\[
\frac{3}{2} kT = \langle \epsilon \rangle = a \int_0^{\infty} \epsilon^{3/2} e^{-\left(\epsilon/\tau\right)^b} \, d\epsilon
\]  

(16)

Combining equations (15) and (16) produces
Energy parameter, $b$

![Figure 1. Effect of energy parameter $b$ on ratio of Maxwellian temperature to kinetic temperature.](image1)

**Figure 1.** Effect of energy parameter $b$ on ratio of Maxwellian temperature to kinetic temperature.

![Figure 2. Energy distribution function. Temperature $kT$, 1 electron volt.](image2)

**Figure 2.** Energy distribution function. Temperature $kT$, 1 electron volt.

\[ kT = \frac{2b \int_0^\infty e^{-(\epsilon / \tau)^b} \epsilon^{3/2} d\epsilon}{3\tau^{3/2} \Gamma(\frac{3}{2b})} = \frac{2}{3} \frac{\Gamma\left(\frac{5}{2b}\right)}{\Gamma\left(\frac{3}{2b}\right)} \]  

(17)

This method defines an electron kinetic temperature that is related to the average energy of the electrons in the same manner as for a Maxwellian distribution. This relation is given in figure 1 for the range of values of $b$ considered herein, while figure 2 shows the general shape of the distribution function plotted for two values of $b$ at a value of $kT$ of 1 electron volt.

**RESULTS AND DISCUSSION**

The rate coefficients were evaluated for different forms of the electron energy distribution function by numerical integration on an IBM 7094 computer. The values obtained for these rate coefficients were then used to solve the general steady-state equations for the fractional amounts of the various particles present.

The fractional amount of the particles present is presented in figures 3 and 4 for different energy distributions (different values of $b$) as a function of electron temperature. The composition for a Maxwellian energy distribution is given for initial densities of $10^9$ and $10^{13}$ particles per cubic centimeter in figure 3. The higher density case (fig. 3(b)) was solved both with and without the inclusion of collisional recombination. This term has little effect in the final results. Neglecting this process had the advantage of simplifying the computer program consider-
ably and was done for all the lower density cases (figs. 3(a) and 4).

The effect of changing the form of the electron energy distribution is illustrated in figure 4, in which the fractional amounts of the different particles are given as a function of electron temperatures for four energy distributions. Figure 4(c) shows that a Maxwellian plasma would be fully ionized for a temperature of 2.5 electron volts, while the fractional amount of atomic ions for a Druyvesteyn plasma would be 0.57. The latter plasma would be completely ionized at temperatures above 3.85 electron volts.

Figure 4 also shows that the type of energy distribution function does not change the shape of a given curve to any extent but merely causes a shift of the curve along the temperature axis. For a given temperature, this figure also shows that the fractional amounts of a given particle can change by orders of magnitude as the distribution function varies. The region where most changes in ionization take place lies between 0.5 and 3.0 electron volts. In this region, the energy distribution of the electrons is as important as the electron temperature in determining the degree of ionization.

In practice, it is doubtful whether the energy distribution will be sufficiently well known to be of use in determining percentage of ionization. With an independent measurement of the percentage of ionization and the electron temperature, however, an
Figure 4. - Fractional amount of molecular neutrals, atomic neutrals, and atomic ions for different forms of distribution function. Original number of particles per cubic centimeter, $10^9$. 

(a) Molecular neutrals. 
(b) Atomic neutrals. 
(c) Atomic ions.
### Table I - Effect of Changes in Rate Coefficients on Report Results

[Original number of particles per cubic centimeter $n_m$, $10^9$; energy parameter $b$, 1; rate coefficient $C_2$, 0.]

<table>
<thead>
<tr>
<th>Kinetic Temperature, $\tau$, eV</th>
<th>Rate Coefficients</th>
<th>Fractional Amounts</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_i$ sec$^{-1}$</td>
<td>$B_i$ sec$^{-1}$</td>
<td>$C_{i'}$ sec$^{-1}$</td>
</tr>
<tr>
<td>0.8</td>
<td>7.940x10$^{-35}$</td>
<td>2.780x10$^{-34}$</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>1.0</td>
<td>3.72x10$^{-33}$</td>
<td>1.28x10$^{-32}$</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>1.5</td>
<td>5.77x10$^{-31}$</td>
<td>2.86x10$^{-30}$</td>
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could easily lead to extremely large errors in predicted plasma composition.

An idea of the sensitivity of the method to the values of the various cross sections used was obtained by changing the values of the rate coefficients used by factors of 0.1 and 10. These data are presented in table I for different values of the kinetic temperature. The first row for each of the three different values of \( \tau \) gives the results that were presented in figure 3(a). The other rows show the effect on the results when various rate coefficients are changed. Radiative (c1) and three-body (f2) recombinations are seen to be the most important.

CONCLUDING REMARKS

The composition of a plasma in which wall processes can be neglected was calculated for various nonthermal energy distributions. The method consisted of deriving a set of steady-state equations that depended on the electron energy distribution function along with appropriate cross sections or collision frequencies.

Figure 5. Comparison of fractional amounts of atomic particles from three different calculations. Number of particles per cubic centimeter, \( n_m \times 10^{13} \).

"Effective" \( b \) can be obtained to describe the electron distribution.

A comparison of the present work (fig. 5) with the Saha equation \( (n = 10^{13}) \) and with the results of Gross and Eisen \( (n = 2.7 \times 10^{13}; \text{ref. } 3) \) shows that a higher temperature is needed to obtain complete ionization than is predicted from their thermodynamic approach. An exact comparison cannot be made with reference 4 since the density was not specified.

The model used herein is believed to be a valid approximation for a well-confined plasma that is formed by nonthermal means. Such a plasma could be formed by a radio frequency electrodeless discharge in a strong magnetic field. For such a case, the use of an equation derived for a plasma in thermodynamic equilibrium
The reactions that were found to be important in determining the composition of the plasma were

1. Electron bombardment of atoms to form atomic ions
2. Electron bombardment of molecules to form molecular ions
3. Radiative recombination of atomic ions
4. Dissociative recombination of molecular ions
5. Dissociation of molecular ions
6. Three-body recombination of atoms to form molecules

The temperature needed to dissociate and ionize the gas completely increased as the distribution was made more non-Maxwellian. A prediction of plasma composition for a given temperature could easily differ by large values if the plasma were non-Maxwellian instead of Maxwellian. The type of energy distribution did not change the shape of any of the curves but only shifted them toward higher temperatures.

All the distribution functions considered predicted complete ionization when the temperature was above 3.85 electron volts.

An equation derived from a thermodynamic approach can lead to large errors in predicting the composition of a nonthermal plasma.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, March 21, 1966.
APPENDIX - EVALUATION OF RATE COEFFICIENTS

From equation (1),

\[ A = \frac{1}{a n_e} \int_0^\infty \sigma_{ai} v \, d n_e \]  
(A1)

The value of \( \sigma_{ai} \) as a function of energy was taken from reference 6 (p. 434) and is given in figure 6. Using this cross section along with equations (14) and (15) for \( d n_e \) and \( a \) allowed the coefficient to be evaluated.

From equation (2)

\[ B = \frac{1}{a \beta n_e} \int_0^\infty \nu_{mo} \, d n_e \]  
(A2)

Values of \( \nu_{mo} \) were obtained from reference 7 and are plotted in figure 7.

The recombination of an electron with an atomic ion can proceed either by radiative or collisional recombination processes (c1) and (c2). These two cases are treated separately.

Little experimental work has been done on atomic radiative recombination coefficients. The experimental data of references 8 and 9 appear to justify the use of the theoretical formulation given by Dewan (ref. 2), wherein

\[ \sigma_{ar} = \frac{1.6 \times 10^{-32}}{\langle e \rangle} \]

Use of this equation gives

![Figure 6. Ionization cross section for atomic hydrogen (ref. 6).](image-url)
A combined radiative-collisional recombination coefficient is treated in reference 10. This combined coefficient is shown to approach a pure radiative recombination coefficient at low densities and become collision dominated at higher densities. The data presented were used to infer that $\alpha_{rc} = \alpha_1 + \alpha_2 n_e$ and that

$$
C_1 = \frac{\langle \alpha_1 \rangle}{a} = \frac{\sigma a r v}{a} = \frac{\sigma a r}{a} \left[ \frac{2 \langle \epsilon \rangle}{m} \right]^{1/2} = \frac{7.51 \times 10^{-19}}{b} \Gamma \left( \frac{3}{2b} \right)
$$

(A3)

Hence,

$$
\langle \alpha_2 \rangle = \frac{1.27 \times 10^{-86}}{\langle \epsilon \rangle^5}
$$

From equation (4) and (A5)

$$
D = \frac{1}{a n_e} \int_0^\infty \sigma_{dr} v \, dn_e
$$

(A5)

The theoretical calculation of the recombination coefficient for dissociative recombination was made by Bates (ref. 11) who found that

$$
\alpha_{dr} = \frac{\text{Constant}}{\epsilon^{1/2}}
$$

(A6)

1A three-body recombination coefficient could have been derived independently of Bates (ref. 10) by using the principle of detailed balancing. (See, e.g., J. B. Hasted, Physics of Atom Collisions, Butterworth & Co., 1964, or Hans R. Griem, Plasma Spectroscopy, McGraw-Hill, Pub. Co., 1964.) However, the method used was deemed sufficient for the purposes of this report. It may be objected that the use of a recombination coefficient derived from Bates is inconsistent with the neglect of excited states in setting up the rate equations. Strickly speaking, this is true. Since the expressions are derived for an optically thin plasma, however, the effect of excited states should be negligible.
An order-of-magnitude-type calculation of the constant gave Bates a value of \( \alpha_{dr} = 10^{-7} \) cubic centimeter per ion second for an electron temperature 250\(^{0}\) K. Biodi and Brown (ref. 12) found an experimental value of \( \alpha_{dr} = 2.5 \times 10^{-6} \) cubic centimeter per ion second for the electron-ion recombination coefficient at a temperature of 300\(^{0}\) K but were not sure of their electron loss mechanism. The cross-sectional data of Rose and Clark (ref. 13) gave a value of the recombination coefficient near that given in reference 11. Their value along with equation (A6) gives

\[
D = \frac{1}{a n_e} \int_0^{\infty} \alpha_{dr} dn_e = \text{Constant} \times \int_0^{\infty} e^{-\left(\varepsilon/\tau\right)^b} d\varepsilon = 2.22 \times 10^{-8} \frac{\tau}{b} \Gamma \left(\frac{1}{b}\right)
\]

This rate coefficient is not strongly dependent on the form of the energy distribution function. From equation (5),

\[
E = \frac{1}{a n_e} \int_0^{\infty} \sigma_{id} \varepsilon dn_e
\]  

Experimental values of \( \sigma_{id} \) could not be found in the literature. Ivash (ref. 14) performed a theoretical calculation of \( \sigma_{id} \) and included the effects of exchange interactions. These interactions affect the cross sections only at energies below 50 electron volts, while for higher energies, \( \sigma_{id} \) may be expressed by

\[
\sigma_{id} \approx \frac{10.75}{\varepsilon^2} \ln \frac{0.1976 \varepsilon}{1.6 \times 10^{-12}} - 0.917
\]

Ivash calculated separate cross sections when a prior interaction or postinteraction was assumed. The values found by assuming postinteraction seemed too high from energy considerations; hence, the prior interaction values were used for energies less than 71 electron volts (fig. 8), while equation (A8) was used for higher energies.

From equation (6),

\[
F = F_1 n_{mo} + F_2 n_{ao}
\]

Reference 15 gives reaction rates for the two three-body processes considered. The values from reference 15 give

\[
F = 2.76 \times 10^{-32} n_{mo} + 9.25 \times 10^{-30} n_{ao}
\]
REFERENCES


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