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Transport coefficients of a chemically reacting plasma

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The analysis for the calculation of the transport coefficients of a chemically reacting partially ionized gas in the presence of a magnetic field is presented here. Taking the phenomenological kinetic equations and considering the ionization and recombination processes only due to electron impact parameter, the Chapman–Enskog method is used to obtain these transport coefficients. Sample numerical calculations are made for the transport coefficients of a potassium-seeded argon gas.

I. INTRODUCTION

Increasing applications of magnetohydrodynamics (MHD) in many engineering devices have made it imperative to study the transport phenomena of a partially ionized gas under different nonequilibrium conditions. As it is difficult to make experiments to measure the transport coefficients like electrical conductivity, viscosity, and thermal conductivity, with any desired accuracy at higher temperatures, mainly theoretical work has been done to study and calculate these coefficients in the presence of cross electric and magnetic fields. But, in most of these papers, the effect of ionizational nonequilibrium is neglected, invoking the condition that the average electron-electron collision frequency is much higher than the inelastic electron collision frequency and that the magnitude of the inelastic gain term is less than the loss term. But, in a partially ionized gas, inelastic collisions are dominant and it is necessary to take account of the effects of ionizational nonequilibrium on the transport phenomena.

During the ionizational nonequilibrium process, the inelastic collisions between the neutral particles of the gas and the charged particles play an important role. This involves energy transfer process between the gas species. The changes in the number density and the temperature of the neutral and the charged particles are determined by conservation laws of charge, energy, etc. The chemical reactions giving rise to ionizational nonequilibrium process are assumed to be related to the modified form of the Saha equation as given by Baum and Fang.

In the present analysis, making use of Baum and Fang’s model of kinetic equations, the electron number densities are evaluated as a solution of continuity and rate equations during ionizational nonequilibrium processes, instead of the Saha equation. Making proper modifications in the energy conservation equation to take account of finite rates of ionization, it is found that the electrons get redistributed and thus as a result of chemical reactions (of ionization-recombination type), the gas will have more electrons at a reduced energy level.

This analysis is then used to obtain the transport coefficients for such a chemically reacting gas assuming that the electron impacts are the only ionization-recombination processes, without distinguishing between the level of states of atoms or ions. The electron number density and electron temperature immediately after the chemical reaction are used in the continuity and energy equations.

The modified mathematical expressions of the transport coefficients due to the effects of chemical reactions are given in Sec. II. These have been derived following the analysis of Mitchner and Kruger In Sec. III, the chemical equilibrium parameters and the method for the evaluation of $n_e$ and $T_e$ from continuity and energy equations are discussed. Section IV gives the numerical results and discussion of the behavior of transport coefficients in a chemically reacting plasma.

II. ANALYSIS

The general form of the kinetic equation as given by Baum and Fang is

$$\left( \frac{\partial}{\partial t} + V \cdot \frac{\partial}{\partial x} + \frac{F_a}{m_a} \cdot \frac{\partial}{\partial V} \right) (n_a f_a)$$

$$= \sum_{a' \alpha} K_{aa'} (n_a \phi_{aa'} - n_a f_a) + a_{a'}(n_a \phi_{a'a} - n_a f_a),$$

where $\phi_{aa'}$ denotes the equilibrium distribution function of the species $a$ and $a'(a, a' = e, i, a)$ in the absence of chemical reactions and $\phi_{a'}$ denotes the distribution function if all the particles were immediately thermalized in each inelastic collision. $K_{aa'}$ is the scattering function and $a_{aa'}$ denotes inelastic collision frequency. The quantities $\phi_{aa'}$ and $K_{aa'}$ are determined using the conservation and irreversibility conditions.

Using the above equation, the effects of chemical reactions on transport coefficients are studied.

Following the analysis of Mitchner and Kruger,1 peculiar particle velocity $C$ and mean mass velocity $U$ are introduced. They satisfy the relation

$$V = C + U.$$

Equation (1) can then be written as

$$\frac{\partial}{\partial t}(n_a f_a) = R'_a + R'_{a'},$$

where

$$\frac{\partial}{\partial t} \left[ \frac{\partial}{\partial x} + \left( U + C_{a'} + \frac{m_a}{m_{a'}} \right) \frac{\partial}{\partial x} \right]$$

and

$$+ \left( \frac{F_a}{m_a} \frac{\partial}{\partial C_{a'}} - C_{a'} \frac{\partial}{\partial C_{a'}} \frac{\partial}{\partial x} \right)$$
For a small departure from the equilibrium, \( f_a(C) \) is written as a cartesian tensor expansion in terms of a peculiar particle velocity

\[
f_a(C) = f_{a(0)}^{(0)}(C) + f_{a(1)}^{(0)}(C) \frac{C_a}{C} + f_{a(2)}^{(0)}(C) \frac{C_B C_a}{C^2} + ... .
\]

(3)

Using Eq. (3) in Eq. (2) and integrating over the differential solid angle \( d \Omega_C \) in velocity space after multiplying by \( C/C \) and \( CC/C^2 \), the equations in terms of cartesian tensor expansion are obtained. Shkarofsky et al.\(^\text{7}\) have given the detailed derivation of these equations but in the absence of inelastic collisions. The equations in terms of the cartesian tensor expansion are written as

\[
\frac{C}{3} \left( \nabla \cdot f_a^{(1)} - \nabla \cdot \frac{df_{a(0)}^{(0)}}{dC} \right) - \frac{e_a}{3m_a} \frac{d}{dC} \left( C \cdot \frac{\partial f_{a(0)}^{(0)}}{\partial C} \right) = \frac{R_a^{(0)}}{1} \nabla \cdot f_a^{(1)} - \frac{R_a^{(1)}}{1} \nabla \cdot f_a^{(2)} - ... .
\]

(4)

\[
f_a^{(0)}(C) = \left( \frac{5}{2} - z^2 \right) \nabla \ln T_a - \frac{e_a}{kT_a} d_a + \omega \times f_a^{(1)} - \frac{1}{R_a^{(0)}} \omega \times f_a^{(1)} - \frac{1}{R_a^{(1)}} \omega \times f_a^{(2)} - \frac{1}{R_a^{(2)}} \omega \times f_a^{(3)} - ...
\]

(5)

where

\[
Z_a = \frac{m_a C}{2kT_a}, \quad w_a = \frac{e_a B}{m_a}, \quad d_a = E', \quad \text{and}
\]

\[
S = \nabla U + \left( \nabla U \right)^T - \frac{1}{2} \omega \left( \nabla U \right).
\]

Since the driving terms \( \nabla \ln T_a, d_a, \) and \( S_a \) for translational nonequilibrium appear linearly and are mutually independent, \( f_a^{(1)}(C), f_a^{(2)}(C) \) can also be written as

\[
f_a^{(1)}(C) = C f_a^{(0)}(C) \left( A_1 \nabla \cdot 1 + A_2 \nabla \cdot 2 + A_3 \frac{B}{B} \times \nabla \right) \ln T_a - \frac{e_a}{kT_a} \left( D_1 d_a + D_2 d_a + D_3 \frac{B}{B} \times d_a \right),
\]

(7)

\[
f_a^{(2)}(C) = C^2 f_a^{(0)}(C) \left( D_1 S_1 + D_2 S_2 + D_3 \frac{B}{B} \times S_a \right),
\]

(8)

where the subscripts 1, 2, and 3 denote the parallel, perpendicular, and Hall components.

\( f_a^{(0)} \) as influenced by chemical reactions is obtained from Eq. (4) following the analysis of Shaw et al.\(^\text{5}\) Thus, \( f_a^{(0)}(C) \) is expanded in terms of Sonine polynomials to evaluate integrals in Eqs. (10)–(13). The resulting expressions for transport coefficients are obtained as

\[
C_\sigma^{(i)} = \left( \frac{n_a}{m_a} + L \right) \left[ e_a^i B^{(i)}, \epsilon k e_a \gamma_1^i, \frac{k}{2} t_2^2 T_a \gamma_2^i \right],
\]

and

\[
C_\sigma^{(i)} = -4 \frac{n_a}{m_a} k T_a \beta^{(i)},
\]

(14)

where

\[
L = H / e^{5/2},
\]

\[
C_{\sigma}^{(i)} = \beta^{(i)} + i \epsilon \gamma^{(i)}...
\]

The expansion coefficients \( \beta^{(i)}, \beta^{(i)}, \gamma^{(i)} \) are obtained by using \( f_a^{(1)}, f_a^{(2)} \) as given in Eqs. (7) and (8):

\[
\sum_{j=1}^{i} \gamma_j e_a \gamma_{a} = \frac{1}{2} (1 + L) \delta_{ij},
\]

(15)

\[
\sum_{j=1}^{i} \beta_j e_a \gamma_{a} = \frac{1}{2} (1 + L) \delta_{ij},
\]

(16)
These equations are solved with the initial conditions

\[ n_e = n_{e0} \]
\[ T_e = T_{e0} \]

at \( t = 0 \). 

where \( F = \frac{p_K}{p_A} \) (ratio of the partial pressure of seed and parent gas),

\[ P_s = p_K + p_A \] is gas pressure,

and

\[ \kappa(T_e) = \left( \frac{2m_e k T_e}{h^2} \right)^{3/2} \exp \left( -\frac{I}{k T_e} \right). \]

With these values of the electron and neutral particle number densities and temperatures, the particle number densities \( n_e, n_s \), and the gas temperature immediately after the chemical reactions are calculated using Eqs. (23)–(27) of Baum and Fang. These equations can be simplified as

\[ n_e = \frac{k' n_o}{2 n_e} \left[ 1 + \frac{8 n_e^2}{n_o k'} - 1 \right], \]
\[ n_s = n_o - \frac{\beta_e n_s n_e}{\alpha_e} \left[ \frac{n_e}{n_s} - 1 \right], \]
\[ T_e = -\frac{2 T_s - 2 T_e^* + \frac{3}{2} I (k/n_e)}{n_e/n_s}, \]
\[ I = k T_e \left[ \log \left( \frac{n_e \times 2.308 \times 10^6 \times T_e^{3/2}}{n_e^2} \right) \right]. \]

These values of \( n_e \) and \( T_e \) are substituted in Eq. (14) to obtain the transport coefficients.

This completes the mathematical formulation and the methodology to calculate the transport coefficient of any gas in the presence of chemical reactions.

IV. NUMERICAL RESULTS AND DISCUSSION

The analytical model developed here is used to make sample numerical calculations and to have a qualitative analysis of the effects of ionizational nonequilibrium on the transport coefficients of argon-potassium plasma. Such a plasma is produced by seeding the argon gas with potassium to have sufficient ionization even at gas temperatures in the range of 2000–2500 °K. This partially ionized gas is used as the working fluid for closed cycle MHD generators. Argon gas has also the advantage of a simple structure and a small collision cross section. In this range of temperature, the calculated values of the transport coefficients can be closely approximated by the use of temperature-independent cross sections, although argon is known to have a very deep Ramsauer minimum in its scattering cross section. To make a comparison with the experimental results of Cool and Zukoski, the cross-section data given by Zukoski et al. is used in the calculations of transport coefficients. These coeffi-
Equilibrium chemically reacting plasma with alkali additives data given by Sakao and Sato.\textsuperscript{10} Ionization and recombination rate coefficients in a nonequilibrium chemically reacting plasma with alkali additives have been investigated theoretically and experimentally by many authors,\textsuperscript{11–14} but their numerical values differ up to a factor of 10\textsuperscript{19}. The commonly used values are also calculated using the more recent cross-section study. These are

\[ \beta_* \approx 1.1 \times 10^{-20} \times T_e^{-4.5} \text{ m}^6/\text{s}. \]

This value also agrees with the value given by Mnatsakanian and Naidis\textsuperscript{14} in the electron temperature range considered here.

To compare the results with the available experimental data\textsuperscript{9,11} the values of other parameters are taken from their study. These are

- Seed percentage = 0.4\% by weight;
- Electron temperature, \( T_e \) = 3000, 3000, 3500 K;
- Gas temperature, \( T_g \) = 2000, 2500, 2500 K;
- Stagnation pressure, \( p \) = 1 atm;
- Electric field, \( E \) = 790 V/m.

Collision cross-sections:

Electron neutral for argon, \( \overline{Q}_{en} = 400 \times 10^{-20} \text{ m}^2 \)

Electron neutral for argon, \( \overline{Q}_{en} = 0.7 \times 10^{-20} \text{ m}^2 \)

from Zukoski et al.\textsuperscript{9};

Electron neutral for argon, \( \overline{Q}_{en} = 230 \times 10^{-20} \text{ m}^2 \)

potassium, \( \overline{Q}_{en} = 0.23 \times 10^{-20} \text{ m}^2 \)

from Sakao and Sato.\textsuperscript{10}

Using the initial values as given above, \( n_e^* \) and \( T_e^* \) during ionizational nonequilibrium processes are calculated from Eqs. (23)–(28). \( n_e^* \) and \( T_e^* \) are obtained at every stage of chemical reactions until it reaches chemical equilibrium. These values are shown in Table I. As a result of ionizational nonequilibrium, electron number density \( n_e^* \) increases, electron temperature \( T_e^* \) decreases and the gas temperature increases. But it may be observed that \( T_e^* \) decreases very slowly as compared to the rate at which \( n_e^* \) increases. Cool and Zukoski\textsuperscript{11} verified that the \( n_e^* \) increases about two orders of magnitude. In their electron temperature measurements at lower relaxation times, they have found that \( T_e \) goes below its initial value but the decrease is very slow. The percent deviation in the initial and final values is given in Table I for comparison. With these values of \( n_e^* \) and \( T_e^* \), first-order nonlinear coupled equations (29) and (30) for \( n_e \) and \( T_e \) are then solved using the fourth-order Runge–Kutta method. Figure 1 gives the variation of \( n_e \) during the ionizational nonequilibrium process for the above-mentioned parameters. As seen from the figure, the ionizational relaxation is completed in time much shorter than the characteristic time of the following plasma. The time scale is in the range of 0.005–1.0 \( \mu \text{s} \).

The successive approximations of the transport coefficients are calculated using Eq. (14) for \( T_e = 3000 \text{ K} \) and \( T_g = 2000 \text{ K} \). With the above values of \( n_e \) and \( T_e \), it was found that convergence increases rapidly as the order of approximation increases, whereas the first approximation differs significantly from the second. There is very little difference between second and third approximations and even this small difference is seen only for a small initial relaxation time. Hence, except for electrical conductivity, coefficients are calculated only up to second approximation.

Figure 2 shows the variation of the normalized electrical conductivity \( \sigma \) during relaxation time. The normalization has been done with respect to its equilibrium value. In these calculations, the average cross-section data given by Zu-

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|}
\hline
Initial value & \( T_e \) & \( T_g \) & \( n_e \) & \( n_e^* \) \\
\hline
Initial value & 3000.00 & 2000.00 & 53.680 & 3.67 \\
Final value & 2821.00 & 2006.10 & 213.40 & 3.667 \\
Percent deviation & 5.97 & 0.305 & 297.540 & 0.082 \\
Initial value & 3000.00 & 2500.00 & 48.13 & 2.94 \\
Final value & 2820.45 & 2504.12 & 203.12 & 2.93 \\
Percent deviation & 5.985 & 0.16 & 326.18 & 0.34 \\
Initial value & 3500.00 & 2500.00 & 169.15 & 2.934 \\
Final value & 3394.43 & 2502.90 & 1125.32 & 2.91 \\
Percent deviation & 3.016 & 0.12 & 565.28 & 0.82 \\
\hline
\end{tabular}
\caption{Initial and final (after chemical equilibrium is reached) values of number densities and temperatures for argon seeded with 0.4\% (by weight) of potassium at 1-atm pressure.}
\end{table}
TABLE II. Comparison of first and second approximations to transport coefficients of argon seeded with 0.4% (by weight) of potassium at 1-atm pressure.

<table>
<thead>
<tr>
<th></th>
<th>First approximation</th>
<th>Second approximation</th>
<th>Percent deviation in first and second approximations</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Case A)</td>
<td>(Case B)</td>
<td>(Case A)</td>
<td>(Case B)</td>
</tr>
<tr>
<td><strong>Electrical conductivity:</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Initial value</td>
<td>248.07</td>
<td>286.70</td>
<td>15.57</td>
</tr>
<tr>
<td>Final value</td>
<td>394.86</td>
<td>729.30</td>
<td>93.14</td>
</tr>
<tr>
<td>Percent deviation of (\sigma) between initial value and final value</td>
<td>59.17</td>
<td>154.40</td>
<td></td>
</tr>
<tr>
<td><strong>Thermal conductivity (\times 10^2):</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Initial value</td>
<td>15.51</td>
<td>16.17</td>
<td>4.26</td>
</tr>
<tr>
<td>Final value</td>
<td>15.97</td>
<td>21.22</td>
<td></td>
</tr>
<tr>
<td>Percent deviation of (\lambda) between initial value and final value</td>
<td>2.93</td>
<td>2.57</td>
<td>4.06</td>
</tr>
<tr>
<td><strong>Viscosity (\times 10^3):</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Initial value</td>
<td>28.24</td>
<td>29.99</td>
<td>6.2</td>
</tr>
<tr>
<td>Final value</td>
<td>28.23</td>
<td>29.91</td>
<td></td>
</tr>
<tr>
<td>Percent deviation of (\eta) between initial value and final value</td>
<td>0.03</td>
<td>0.01</td>
<td>0.003</td>
</tr>
</tbody>
</table>

koski et al.\(^9\) [Case (A)] and Sakao and Sato\(^10\) [Case (B)] are used. These curves are represented by continuous lines for Case (A) and by broken lines for Case (B). The first three approximations of the electrical conductivity are computed to examine the rate of convergence of these approximations. The second and third approximations are found to converge, but as seen from the figure the convergence is not very fast at lower relaxation times. This is true for both cases of the collision cross sections considered here. Surprisingly, for higher relaxation times, the difference in the values of the conductivity for Case (A) and Case (B) is very small (for both the second and third approximations). The measured values of electrical conductivity given by Cool and Zukoski\(^11\) are shown in Fig. 2 [Case (A)] and are found to be comparable with the first approximation of the present results. Table II gives the first and second approximations of the initial and the final values and the percent deviation.

In Fig. 3, the variation of thermal conductivity with time is shown. Here only the first (= second) and third approximations of the thermal conductivity are calculated for two different cross-section data—Case (A) and Case (B). Mainly, the electrons and neutral particles contribute to the thermal conductivity. In the present case the electron number density increases due to inelastic collisions during ionization relaxation time and hence thermal conductivity will also increase. Unlike the case of the electrical conductivity, the different cross sections of Case (A) and Case (B) significantly affect the third approximations of the thermal conductivity. This behavior of thermal conductivity is comparable qualitatively with the theoretical calculations of Devoto\(^2\) for pure argon gas.

The behavior of viscosity \(\eta\) in the presence of chemical reactions is shown in Fig. 4. In the calculations of viscosity, mainly heavy particles contribute. Its value increases with the gas temperature but as the ionization begins it starts decreasing. In the presence of chemical reactions in a partially ionized seeded gas, due to inelastic collisions, the degree of ionization increases even while the gas temperature is moderate and nearly remains constant. But this behavior is not seen in the calculation of viscosity for a neutral gas. Devoto\(^2\) and Kannappan and Bose\(^3\) have made theoretical calculations for the viscosity of a pure gas. In their calculations, \(\eta\) increases as temperature increases from 5000 to 10 000 K, as the degree of ionization in this range is negligible. But as the temperature is increased further, the degree of ionization also increases and then the viscosity starts decreasing. In the present analysis, a seeded gas is considered, where the ion-
zation due to inelastic collisions increases while the temperature remains almost same. Hence, the viscosity is nearly constant but shows a decreasing trend as the ionization increases with relaxation time. This behavior of viscosity can be seen more clearly for higher temperatures, where the degree of ionization is more.

Thus, it is concluded that chemical reactions affect transport coefficients significantly. As is seen from Table II, the electrical conductivity is enhanced substantially from its thermal equilibrium value, the thermal conductivity is increased modestly and the viscosity is reduced but negligibly.

ACKNOWLEDGMENT

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APPENDIX

<table>
<thead>
<tr>
<th>$a_1$</th>
<th>$a_2$</th>
<th>$a_3$</th>
<th>$a_4$</th>
<th>$a_5$</th>
<th>$a_6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>1.5</td>
<td>1.875</td>
<td>4.3125</td>
<td>6.7656</td>
<td></td>
</tr>
</tbody>
</table>

\[ \nu_{el} = a_1 + a_2 + a_3 + a_4 + a_5 + a_6 \]

\[ \nu_{ea} = a_7 + a_8 - 0.5 a_9 - 0.125 \]

\[ a_9 - 0.5 a_10 + 3.25 a_11 - 1.4375 \]

\[ a_9 - 0.125 a_11 - 1.4375 a_12 + 6.7656 \]

\[ a_{13} + 0.5 a_{14} + 0.0 a_{15} + 0.0 \]

\[ a_{14} + 0.0 a_{16} + 3.75 a_{17} + 0.0 \]

\[ a_{15} + 0.0 a_{17} + 0.0 a_{18} + 2.8125 \]

where $a_i$ to $a_{18}$ are given by

\[ a_1 = H/\epsilon, \]

\[ a_2 = (0.5 - 3.0/\epsilon) H/\epsilon, \]

\[ a_3 = (1.375 - 6.5/\epsilon + 1.0/\epsilon^2) H/\epsilon, \]

\[ a_4 = (-5.75 + 5.0/\epsilon - 2.0/\epsilon^2) H/\epsilon, \]

\[ a_5 = (10.3125 - 24.375/\epsilon + 4.5/\epsilon^2) H/\epsilon, \]

\[ a_6 = (25.8906 - 0.3906/\epsilon + 0.250/\epsilon^2) H/\epsilon, \]

\[ a_7 = 4.0 Q, \]

\[ a_8 = (-11.0 - 5.0/\epsilon - 1.0/\epsilon^2) Q, \]

\[ a_9 = (10.0 - 4.5/\epsilon + 6.0/\epsilon^2) Q, \]

\[ a_{10} = (33.0 - 27.0/\epsilon + 12.0/\epsilon^2) Q, \]

\[ a_{11} = (-27.375 + 61.125/\epsilon - 33.0/\epsilon^2) Q, \]

\[ a_{12} = (18.6875 - 71.875/\epsilon + 49.5/\epsilon^2) Q, \]

\[ a_{13} = 9.125 R, \]

\[ a_{14} = 13.75(-1.0 + 1.0/\epsilon) R, \]

\[ a_{15} = 5.625(-1.0 + 1.0/\epsilon^2) R, \]

\[ a_{16} = (15.75 - 20.25/\epsilon + 11.25/\epsilon^2) R, \]

\[ a_{17} = (-11.25 + 33.75/\epsilon - 22.5/\epsilon^2) R, \]

\[ a_{18} = (5.625 - 11.25/\epsilon + 22.5/\epsilon^2) R, \]

where

\[ Q = 2.0 H/\epsilon^2 \quad \text{and} \quad R = 0.75\sqrt{\tau H/\epsilon^{12}}. \]

$\nu_{el}$, $\nu_{ea}$, and $\nu_{\omega}$ are the mean $e-i$, $e-a$, and $a-a$ particle collision frequencies.