Thermodynamic and Transport Properties of Real Air Plasma in Wide Range of Temperature and Pressure*

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Abstract Air plasma has been widely applied in industrial manufacture. In this paper, both dry and humid air plasma’s thermodynamic and transport properties are calculated in temperature 300–100000 K and pressure 0.1–100 atm. To build a more precise model of real air plasma, over 70 species are considered for composition. Two different methods, the Gibbs free energy minimization method and the mass action law method, are used to determine the composition of the air plasma in a different temperature range. For the transport coefficients, the simplified Chapman-Enskog method developed by Devoto has been applied using the most recent collision integrals. It is found that the presence of CO2 has almost no effect on the properties of air plasma. The influence of H2O can be ignored except in low pressure air plasma, in which the saturated vapor pressure is relatively high. The results will serve as credible inputs for computational simulation of air plasma.

Keywords: air plasma, thermodynamic properties, transport coefficients

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(Some figures may appear in colour only in the online journal)

1 Introduction

Air plasma has been widely applied in industrial manufacturing, including cutting, welding, spraying, metallurgy, waste destruction and surface treatment. It is also commonly used in electrical equipment, especially in medium-voltage circuit breaking. Besides, air can also permeate into any plasma which performs in open atmosphere [1]. Due to the complicated and changing application environment, the study of air plasma in a wide temperature and pressure range is necessary. Much of the previous research has focused on numerical modelling of air plasma [2–4], which is an essential method of air plasma study.

Reliable physical models of arc plasma require accurate thermodynamic and transport properties. Murphy et al. [1] calculated the transport properties of a N2-O2 mixture with considering CO2 in atmospheric pressure and indicated the significant influence of CO2 on the viscosity of air plasma. Capitelli et al. [5] presented properties of N2-O2 mixture for a wide pressure range (0.01–100 atm) and temperature range (50–100000 K). Wang et al. [6] calculated the properties of a N2-O2 mixture plasma with consideration of argon as a replacement of the other composition of real air. Angola et al. [7] calculated the properties of a N2-O2 mixture plasma with the phenomenological potential model. Kagoné et al. [8] researched the transport properties of air-water mixtures. However, to the best of our knowledge, no data have been presented for real air plasma, with consideration of N2, O2, Ar, CO2 and H2O for a wide temperature and pressure range.

The collision integral is one of the prerequisites for transport coefficients calculation. The application of different intermolecular potentials, from which the collision integral is derived, can make a discrepancy in different calculation results. In this work, following the recommendation of Angola [7], we used the phenomenological potential developed by Cambi et al. [9], which significantly improved the reliability of our calculation.

In this paper, the thermophysical properties of air, with consideration of the influence of CO2 and H2O, have been calculated for a wide temperature range (300–100000 K) and a wide pressure range (0.1–100 atm). The calculation is under the LTE (local thermodynamic equilibrium) and LCE (local chemical equilibrium) assumption, which is reasonable and widely applied in the arc plasma simulation under this temperature and pressure condition. The influence of H2O proportions especially in low pressure is discussed.

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2 Method of calculation

2.1 Composition and thermodynamic properties

The calculation of thermodynamic and transport properties was based on the composition. Totally 71 different species have been considered, listed in Table 1. Two different methods were adopted to calculate the air plasma composition under the assumption of LTE and LCE [10]. For temperature below 50 000 K, the species composition of air was obtained based on the minimization of Gibbs free energy. All the data needed for this calculation were obtained from Refs. [11]–[13]. For temperature higher than 50 000 K, where only ionization reactions happen, the law of mass action was applied to determine the plasma composition. The data needed for partition function calculation were taken from the energy levels data tabulated by Moore [14] and the most recent data provided by the National Institute of Standards and Technology (NIST) [15]. These two methods overlapped between 40 000 K and 50 000 K giving a link zone to validate the consistency. The Debye-Hückel correction has been taken into account in the Dalton laws and partition function calculation [16].

In the present work, the thermophysical properties of both dry air and humid air were calculated. The dry air consisted of 78% N2, 21% O2, 0.97% Ar and 0.03% CO2 by mole fraction. For humid air, the ratio among N2, O2, Ar and CO2 was the same as dry air. The maximal proportion of H2O in air can be estimated by saturated vapor pressure $P_s$ using Antoine’s equation, which only depends on temperature $T$

$$\log_{10} P_s = A - \frac{B}{C + T}.$$  \hspace{1cm} (1)

The $A$, $B$ and $C$ in Eq. (1) were constants. For water vapor $A$ is 8.07131, $B$ is 1730.63 and $C$ is 233.426 (when $T$ is 0°C–100°C).

Calculation of the partition function is a prerequisite for calculating the plasma composition. For monatomic species, the internal partition function only consists of the electronic contribution and is given by

$$Z_i = Z_i^e(T_e) = \sum_n g_n \exp\left(-\frac{\varepsilon_n}{kT_e}\right),$$  \hspace{1cm} (2)

where $Z_i^e(T_e)$ is the electron internal partition function of species $i$, $g_n$ and $\varepsilon_n$ are the degeneracy and energy of the $n$th electronic levels, and $E_i$ eff is the effective ionization limit at which the sum over electronic levels is cut off, which is given by [16]

$$E_i \text{ eff} = E_i - \Delta E = E_i - \frac{q^2(z_e + 1)}{4\pi\varepsilon_0\lambda_D}.$$  \hspace{1cm} (3)

The Debye length in the above equation is calculated without consideration of the ion contribution. The data of high energy levels with high principal quantum number, which are not given by Moore or the NIST database, were estimated using the hydrogenic approximation based on the last principal quantum number [17]. The energy states, the associated degeneracy, and the ionization limits used in the present work are taken from Ref. [18].

For diatomic and polyatomic species, the internal partition function is the product of electronic, vibration and rotation partition functions. In the present paper, we use the $T_e$ to describe the excited electron energy and the heavy species temperature $T_h$ to describe the vibrational energy and rotational energy. The expression is given by [19]

$$Z_i = Z_i^e(T_e) Z_i^{\text{vib}}(T_h) Z_i^{\text{rot}}(T_h),$$  \hspace{1cm} (4)

where $Z_i^{\text{vib}}(T_h)$ and $Z_i^{\text{rot}}(T_h)$ are respectively the vibrational and rotational partition functions of species $i$.

The internal partition functions of diatomic species are calculated by the Morse potential minimization method. For polyatomic species, due to the lack of accurate spectroscopic data, the internal partition functions are calculated by a simplified method, in which the discrete summations over rotational quantum numbers are replaced by an integral form [17].

It should be noted that the Virial correction has not been considered in this paper, following the recommendation of Angola [20] and Colonna [21], who indicated that the virial correction is appreciable at high pressures (higher than 200 bar) and low temperature (lower than 1,500 K) as discussed in Ref. [16].

After obtaining the composition and partition function of each species, following the standard thermodynamic formulas, the thermodynamic properties can be evaluated directly.

<table>
<thead>
<tr>
<th>Element</th>
<th>Species</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>H2, H, H+ , H−</td>
</tr>
<tr>
<td>C</td>
<td>C, C2, C5, C7, C− , C+, C2+, C3+, C4+, C5†</td>
</tr>
<tr>
<td>N</td>
<td>N2, N, N3, N5, N+, N2+, N3+, N4+, N5+, N6†</td>
</tr>
<tr>
<td>O</td>
<td>O2, O, O3, O5, O7, O−, O+, O2+, O3+, O4+, O5+, O6†</td>
</tr>
<tr>
<td>Ar</td>
<td>Ar, Ar+, Ar2+, Ar3+, Ar4+, Ar5+, Ar6+, Ar7†</td>
</tr>
<tr>
<td>H, O</td>
<td>H2O, H2O2, OH, OH−, OH+</td>
</tr>
<tr>
<td>O, N</td>
<td>NO, NO2, NO+, NO−, NO2+, NO3+, N2O, N2O−, NO3−</td>
</tr>
<tr>
<td>C, O</td>
<td>CO, CO2, CO+, CO−, CO2+, CO2−, CO3, CO5</td>
</tr>
<tr>
<td>Others</td>
<td>CH, CH+, NH, NH+, HNO, HCO</td>
</tr>
</tbody>
</table>
2.2 Transport coefficients and collision integrals

The Chapman-Enskog method [22] generalized by Hirschfelder et al. [23] was adopted to calculate the transport coefficients. By solving the linear equations introduced by Sonine polynomials, different transport properties can be obtained. To simplify the calculation, the method of Devoto [24,25] and Bonnefoi [26] was used in this work, which was proved to be accurate under the LTE assumption by Colombo et al. [27]. For the thermal conductivity calculation, the theory presented by Butler et al. [28] has been applied. Third order approximation was used for all transport coefficients except the viscosity using the second order approximation.

The bracket integrals, which appear in the transport coefficients calculation, can be represented by collision integrals [23]. The collision integrals between different particles need to be calculated by different methods.

2.2.1 Collision integrals between neutral particles

In the previous works many different potential models have been used in the collision integral calculations between neutral particles. In this paper, we used a new potential model, named phenomenological model, which was developed by Cambi et al. [29] and Capitelli et al. [29,30]. This potential model can give a satisfactory description of both the long-range and short-range interaction potential. An excellent agreement was achieved when comparing the phenomenological model potential with the Aziz two-body potential, which was more accurate but requires quantum mechanical approaches and high computational cost [31].

The parameters of the phenomenological potential was mainly calculated from the polarizability \( \alpha \) of the interaction particles, which was mainly taken from the NIST database [32] and previous estimation and given in Table 2.

<table>
<thead>
<tr>
<th>Species</th>
<th>Polarizability (Å(^3))</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>0.667</td>
</tr>
<tr>
<td>N</td>
<td>1.1</td>
</tr>
<tr>
<td>Ar</td>
<td>1.594</td>
</tr>
<tr>
<td>OH</td>
<td>1.47</td>
</tr>
<tr>
<td>NO</td>
<td>1.7</td>
</tr>
<tr>
<td>H(_2)O</td>
<td>1.45</td>
</tr>
<tr>
<td>CO</td>
<td>1.935</td>
</tr>
<tr>
<td>N(_2)</td>
<td>3.338</td>
</tr>
<tr>
<td>CH</td>
<td>2.158</td>
</tr>
<tr>
<td>O(_3)</td>
<td>2.684</td>
</tr>
<tr>
<td>N(_2), O(_2), NO, N, O</td>
<td>1.7403</td>
</tr>
</tbody>
</table>

Table 2. Polarizability of neutral species

For the cross sections of the interaction between electrons and neutral monatomic particles, the approach recommended by Laricchiuta et al. [35] was used in this work. The required data of cross sections were taken from the database of the Plasma Data Exchange Project [36]. For the interactions between electrons and N\(_2\), O\(_2\), NO, N and O, the collision integrals were taken from numerical results proposed by Capitelli et al [37]. For some electron-neutral collision interactions, whose cross section data cannot be found in the Plasma Data Exchange Project, the polarization potential has been adopted. The data source of electron-neutral collision calculation is given by Table 3.

<table>
<thead>
<tr>
<th>Species</th>
<th>Data source</th>
</tr>
</thead>
<tbody>
<tr>
<td>N(_2), O(_2), NO, N, O</td>
<td>Capitelli et al. [37]</td>
</tr>
<tr>
<td>Ar, C, C(_2), CH, CO, CO(_2), H, H(_2), H(_2)O, NO, NO(_2), O(_3), OH, N(_3), H(_2)O</td>
<td>Plasma data exchange project [36]</td>
</tr>
</tbody>
</table>

2.2.2 Collision integrals between ions and neutral particles

Both the elastic and inelastic collisions process should be considered in the interactions between neutral particles and ions. Following the work of Murphy [33], the total collision integrals were estimated using the empirical mixing rule

\[
\Omega^{(l,s)} = \sqrt{(\Omega_{\text{in}}^{(l,s)})^2 + (\Omega_{\text{el}}^{(l,s)})^2},
\]

where \( \Omega_{\text{in}}^{(l,s)} \) was the inelastic collision integral and \( \Omega_{\text{el}}^{(l,s)} \) was the elastic interactions, which can be obtained by adopting the phenomenological potential [27].

The charge transfer cross section was given by

\[
Q_{\text{ex}} = (A - B \ln E)^2,
\]

where \( A \) and \( B \) were constants obtained from experimental or theoretical methods. \( E \) was the collision energy. The data of N-N\(^+\), O-O\(^+\), C-C\(^+\), Ar-Ar\(^+\) and H-H\(^+\) interactions were taken from the work of Copeland and Crothers [34].

In this work, the inelastic collision integrals between unlike species and collision integrals with even \( l \) were neglected, due to their small value compared to the elastic collision integrals.

2.2.3 Collision integrals between electron and neutral particles

For the cross sections of the interaction between electrons and neutral monatomic particles, the approach recommended by Laricchiuta et al. [35] was used in this work. The required data of cross sections were taken from the database of the Plasma Data Exchange Project [36].

For the interactions between electrons and N\(_2\), O\(_2\), NO, N and O, the collision integrals were taken from numerical results proposed by Capitelli et al [37]. For some electron-neutral collision interactions, whose cross section data cannot be found in the Plasma Data Exchange Project, the polarization potential has been adopted. The data source of electron-neutral collision calculation is given by Table 3.

2.2.4 Collision integrals between charged particles

The screened Coulomb potential was used to calculate the collision integrals of charged-charged interactions. There were some debates on whether the screened effect of ions should be considered when calculating the Debye length. Different definitions of the Debye length can significantly affect the charged-charged interactions.
interactions and further more affect the transport coefficients. Murphy found that the calculated argon electrical conductivity agreed better with the measured value when neglecting the ions in the Debye length calculation. In the present work, we used the method originally presented by Liboff and summarized by Capitelli with the Debye length without consideration of ions.

3 Results and analysis

In this part, the composition, thermodynamic and transport properties of both dry and humid air under the LTE assumption for different temperatures (300–100000 K) and pressures (0.1–100 atm) are presented. The results have been compared with some previously published data and showed good agreement. The influences of temperature, pressure and water proportions on the composition, thermodynamic properties and transport coefficients on air plasma systems are discussed.

3.1 Properties for dry air plasma at atmospheric pressure

In order to validate the data and equations used in the calculation, the properties of dry air plasma, which consists of 78% N\textsubscript{2}, 21% O\textsubscript{2}, 0.97% Ar and 0.03% CO\textsubscript{2}, are presented in the first part of this section, with a comparison with some previous works. Besides, the influences of temperature and the presence of CO\textsubscript{2} are discussed in this part.

The composition of dry air mixture plasma under the LTE condition and atmospheric pressure is shown in Fig. 1 as a function of temperature. The dissociations of CO\textsubscript{2}, O\textsubscript{2}, CO and N\textsubscript{2} occur at 1200 K, 3000 K, 6300 K and 7000 K respectively. The N ionizes in a stepwise manner at around 14000 K while the O successively ionizes at 15000 K. It should be noted that the composition was obtained based on different methods in different temperature ranges. For temperature lower than 50000 K, the minimization Gibbs free energy method was adopted to obtain the composition. For temperature higher than 50000 K the law of mass action was applied. Fig. 1(b) shows a good consistency between the results of two methods in the link temperature zone (40000–50000 K).

The ionization process of dry air, as shown in Fig. 2(a), is mainly dominated by successively ionization of N and O, since they are chemical elements with the highest mole fraction. At around 14000 K, the electron number density increases sharply because of the ionization of N atom and O atom. When temperature is higher than 25000 K, the electron number density tends to be stable as a result of the balance between the ionization reaction and the rarefaction effect, related to the pressure conservation.

As shown in Fig. 2(b), the mass density shows an inverse correlation with the temperature, due to the rarefaction effect and dissociation reaction.

Fig. 1 Temperature dependence of the number density of different species in dry air plasma at atmospheric pressure (a) under 30000 K and (b) 30000–100000 K

Fig. 2 Temperature dependence of (a) ionization process, (b) mass density, (c) Enthalpy and (d) specific heat of dry air plasma in a wide temperature range at atmospheric pressure. The results of Capitelli et al. are shown with symbols for comparison.
Fig. 2(c) and (d) show the specific enthalpy and specific heat, respectively. The peaks in specific heat for temperature higher than 14000 K are mainly because of the successively ionization of N and O. The ionization of N$^{5+}$ and O$^{6+}$ occurs at a relatively high temperature (higher than 100000 K) due to their high ionization energy, which leads to the sharp decreases of specific heat and reactive thermal conductivity at 90000 K.

Fig. 3 shows the viscosity of dry air plasma at atmospheric pressure. The viscosity first increases with temperature due to the $T^{1/2}$ dependence of viscosity and the collision integral between neutral species decreasing. Subsequently the viscosity decreases because the strong Coulomb interaction starts to dominate. Our results agree well with those of Angola et al. [7] but are lower than results of Murphy et al. [1] and Wang et al [6]. This deviation is the caused by a different selection of neutral-neutral interaction potential. The application of phenomenological potential can increase the value of neutral-neutral collision integral and furthermore decrease the viscosity value, as shown in the curves of Angola’s results and ours in Fig. 4. Besides, in our results, the influence of CO$_2$ on the viscosity can be neglected, which is contradictory with the result of Murphy et al., in which the low proportion of CO$_2$ (0.03%) can significantly increase the viscosity by maximal 15% [1]. This discrepancy is also due to the different neutral-neutral collision integral we used. However, our results may be more reasonable due to the usage of recently published intermolecular potential data.

As shown in Fig. 5, the total thermal conductivity ($\lambda_{\text{total}}$) is composed of the translational thermal conductivity of electron ($\lambda_{\text{tra}}$), the translational thermal conductivity of heavy species ($\lambda_{\text{trh}}$), the internal thermal conductivity ($\lambda_{\text{int}}$) and the reactive thermal conductivity ($\lambda_{\text{reac}}$). According to Fig. 5, $\lambda_{\text{total}}$ of dry air is mainly decided by $\lambda_{\text{tra}}$ and $\lambda_{\text{reac}}$ at a temperature under 10000 K, due to the high proportion of neutral molecules in the plasma. For temperature between 10000 K and 20000 K, the ionization process occurs and $\lambda_{\text{total}}$ is affected by both $\lambda_{\text{tra}}$ and $\lambda_{\text{reac}}$. When the temperature is above 20000 K, $\lambda_{\text{total}}$ is completely dominated by component $\lambda_{\text{tra}}$. This tendency is consistent with the previous research on the air plasma, as shown in Fig. 6, except Murphy’s result, which is higher than our result at a temperature around 7000 K. This discrepancy is due to the different values of collision integrals between the neutral particles we used. Besides, the presence of CO$_2$ can slightly increase $\lambda_{\text{total}}$ at temperatures between 10000 K and 20000 K, which is due to the higher value of C-C$^+$ and O-O$^+$ collision integrals.

As shown in Fig. 7, the electrical conductivity increases sharply in the temperature range from 10000 K to 20000 K due to the ionization process. For temperatures higher than 20000 K, the electrical conductivity increases wavyly under the combined influence of ionization and rarefaction effect. Our results show good agreement with the previously published work, as shown in Fig. 8.
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3.2 The influence of pressure on dry air plasma

Fig. 9 shows the properties of dry air at different pressure. The pressure increase shifts the dissociation and ionization reactions to a higher temperature range, according to Chatelier’s principle. The crossing phenomenon of electrical conductivity at around 15000 K is due to the change of main elastic collision involving electrons [41].

3.3 The influence of H$_2$O on humid air plasma

In this section, the influence of H$_2$O on humid air plasma is discussed. In the atmospheric pressure, the presence of H$_2$O has almost no influence on air plasma properties, due to the limited proportion of H$_2$O (less than 4% at room temperature according to Antoine’s equation). However, in low temperature, the maximal proportion of H$_2$O is much larger and the effect is not ignorable.

According to Antoine’s equation and Dalton’s Law of Partial Pressure, the maximal proportion of H$_2$O in air at atmospheric pressure, which can be estimated by the saturated vapor pressure, is 4%. As shown in Fig. 10, at 3000 K and 13000 K, the presence of H$_2$O slightly increases the peaks of the total thermal conductivity because of the dissociation of H$_2$O and the ionization of H, respectively. On the other hand, the presence of H$_2$O has almost no influence on other air plasma properties, due to the limited proportion. Therefore, at atmospheric pressure, the presence of H$_2$O is ignorable.

Fig. 9 Temperature dependence of (a) mass density, (b) enthalpy, (c) specific heat, (d) total thermal conductivity, (e) viscosity and (f) electrical conductivity of dry air plasma at different pressure

Fig. 10 Temperature dependence of total thermal conductivity of both dry and humid air at atmospheric pressure
Fig. 11 shows the influence of H$_2$O on the air plasma properties at low temperature pressure. According to Antoine’s equation and Dalton’s Law of Partial Pressure, the maximal proportion of H$_2$O at a pressure of 0.1 atm is 40%. For specific heat and reactive thermal conductivity, as shown in Fig. 11(a) and (b), the presence of H$_2$O significantly increases the peak values at temperatures around 3000 K and 13000 K, which are respectively caused by H$_2$O dissociation and H ionization. Besides, the presence of H$_2$O also decreases the mole fraction of N and O and therefore decreases the specific heat peak values at 60000 K and 80000 K.

![Fig.11 Temperature dependence of (a) specific heat, (b) reactive thermal conductivity, (c) viscosity and (d) electrical conductivity of air plasma with different H$_2$O proportions at a pressure of 0.1 atm](image)

The presence of H$_2$O also decreases the peak value of the viscosity at around 10000 K as shown in Fig. 11. As explained by Cressault et al. [41], the viscosity is directly proportional to the $M^{1/2}$ where the $M$ is the mass of the particle. Therefore, the lowering of viscosity when the H$_2$O proportion increases is mainly due to the small mass of H.

Besides, the electrical conductivity increases as the H$_2$O proportion at temperatures higher than 20000 K, as shown in Fig. 11(d). This is due to the higher electron mobility in the humid air plasma system.

### 4 Conclusion

In this paper, the thermophysical properties of both dry and humid air plasmas have been presented for temperatures in the range of 300–100000 K and pressures in the range of 0.1–100 atm. The calculation is under the LTE and LCE assumption, which is reasonable and widely applied in the arc plasma simulation under these temperature and pressure conditions. The Debye correction is taken into account in the calculation while the Virial correction has been neglected, which is not necessary under the present calculation condition [16,20,21]. To build a more precise model of real air plasma, over 70 species are considered in the calculation. The most recent data and interaction potential have been used in the calculation of the collision integral. The results will serve as reliable inputs for computational simulations of the behavior of air plasmas in a wide range of temperatures and pressures.

At atmospheric pressure, the air plasma is mainly dominated by dissociation reactions at temperatures under 10000 K and by successive ionization processes of N and O at temperatures above 14000 K. The ionization of N$^{6+}$ occurs at a relatively high temperature, which leads to the sharp decreases of specific heat and reactive thermal conductivity at around 90000 K.

The present work shows that the presence of CO$_2$ (0.03% by mole fraction) has almost no effect on the properties of air plasma, which is contradictory with the result of Murphy et al. This discrepancy is caused by the different neutral-neutral collision integral we used. However, our results may be more reasonable due to the usage of recently published intermolecular potential data.

At atmospheric pressure, there is little difference between the properties of humid air plasma and dry air plasma, due to the limited proportion of H$_2$O. However, at low pressure, the saturated vapor pressure is relatively high and the presence of H$_2$O cannot be ignored.

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