Effects of copper vapour on thermophysical properties of CO$_2$-N$_2$ plasma

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Received 5 April 2016 / Received in final form 20 June 2016
Published online 3 November 2016 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2016

Abstract. CO$_2$-N$_2$ mixtures are often used as arc quenching medium (to replace SF$_6$) in circuit breakers and shielding gas in arc welding. In such applications, copper vapour resulting from electrode surfaces can modify characteristics of plasmas. This paper therefore presents an investigation of the effects of copper on thermophysical properties of CO$_2$-N$_2$ plasma. The equilibrium compositions, thermodynamic properties (including mass density, specific enthalpy, and specific heat), transport coefficients (including electrical conductivity, viscosity, and thermal conductivity), and four kinds of combined diffusion coefficients due to composition gradients, applied electric fields, temperature gradients, and pressure gradients respectively, were calculated and discussed for CO$_2$-N$_2$ (mixing ratio 7:3) plasma contaminated by different proportions of copper vapour. The significant influences of copper were observed on all the properties of CO$_2$-N$_2$-Cu mixtures. The better ionization ability and larger molar mass of copper and larger collision integrals related to copper, should be responsible for such influences.

1 Introduction

CO$_2$, N$_2$, and their mixtures have been dedicated many efforts to a few decades because of their wide applications both in astrophysics and industries. For example, as main components of many planetary atmospheres (e.g. Martian and Venusian atmospheres), the high-temperature CO$_2$-N$_2$ mixtures have been studied for their thermodynamic and transport properties since 1960s [1,2]. Even recently, the research on CO$_2$-N$_2$ plasma can be found for thermophysics and hydrodynamics [3–8]. In industries, due to the high global warming potential (GWP) of SF$_6$ (24 000 times higher than that of CO$_2$ [9]), CO$_2$ and N$_2$ are also applied to replace SF$_6$ and reduce GWP, for example, as insulating medium in gas-insulated switchgears (GIS) [10] and as arc quenching medium in gas circuit breakers (GCB) [11]. Besides, CO$_2$ and N$_2$ are used as shielding gases in metal-inert-gas (MIG) welding and gas metal arc welding (GMAW) [12,13].

However, few attention was paid to effects of impurities (e.g. metallic vapour) on CO$_2$-N$_2$ plasma. Actually, the presence of such impurities could modify significantly characteristics of plasmas. For example, in a number of arc devices, such as circuit breakers, arc heaters, torches, and arc welding apparatus, the metallic vapour (e.g. copper vapour) resulting from electrode surfaces can mix with a working gas and thus exert influence on arc characteristics [14]. Chervy et al. [15], Paul et al. [16], and Wang et al. [17] all found that electrical conductivity of SF$_6$-Cu mixtures is strongly dependent on copper concentration. Zhong’s et al. [18], Cressault and Gleizes [19], Cressault’s et al. [20] works on combined diffusion coefficients also revealed the significant effect of metallic vapour on SF$_6$ and Air plasmas. In arc welding, the presence of metal vapour was proved to have a major influence on the properties of arc and the size and shape of weld pool [21]. In extinction process of circuit breakers, copper contamination due to electrode erosion was also proved to have a cooling effect at the arc center and broaden the arc column [22,23]. Therefore, the thermophysical properties of CO$_2$-N$_2$ plasma are expected to be affected by metal vapour (e.g. copper in the present investigation) when used as arc quenching gas in GCB or shielding gas in MIG and GMAW. To our knowledge, no corresponding works have been reported. What’s more, the combined diffusion coefficients which were proposed by Murphy [24–27] and greatly simplify the treatment of diffusion, are indispensable input data in developing model of metal vapour transfer both in arc welding [28] and circuit breakers [29]. Another aim of the present work is therefore to provide these four kinds of combined diffusion coefficients due to composition gradients, applied electric fields, temperature gradients, and pressure gradients respectively [18], which have not been reported either.

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In this paper, the influences of copper vapour on thermophysical properties of CO$_2$-N$_2$ plasmas are investigated at temperatures of 2000–30000 K. The collision integrals were calculated using the newly developed potential and cross sections. The properties include equilibrium compositions and thermodynamic properties (mass density, specific enthalpy, and specific heat), transport coefficients (electrical conductivity, viscosity, and thermal conductivity), and four kinds of combined diffusion coefficients (combined ordinary diffusion coefficient, combined electric field diffusion coefficient, combined temperature diffusion coefficient, and combined pressure diffusion coefficient), which are presented and discussed in Sections 2–4, respectively. It is notable that copper and/or copper oxide are likely to condense at low temperatures and hence the plasma is not established in CO$_2$-N$_2$-Cu mixtures but in CO$_2$-N$_2$ mixtures. Therefore, the lower limit of temperature is set to be 2000 K in this work, following the recommendation of Cressault et al. [20,30].

2 Effects of copper on compositions and thermodynamic properties

The particles in CO$_2$-N$_2$-Cu mixtures (e.g. CO$_2$-N$_2$ plasma contaminated by copper vapour) is assumed to be gaseous and in local thermodynamic equilibrium (LTE). The minimization of the Gibbs free energy is then used to obtain the equilibrium compositions, as described in our previous work [14,31–33]. Totally 76 species, namely C, C$_2$, C$_3$, C$_4$, C$_5$, CO, CO$_2$, C$_2$O, C$_3$O$_2$, CN, CN$_2$, C$_2$N$_2$, C$_4$N$_2$, N, N$_2$, N$_3$, NO, NO$_2$, NO$_3$, N$_2$O, N$_2$O$_3$, N$_2$O$_4$, NCO, O, O$_2$, O$_3$, Cu, Cu$_2$, CuO, C$^{+}$, C$_2$$^{+}$, C$_3$$^{+}$, C$_4$$^{+}$, C$^{-}$, C$_2$$^{-}$, C$_3$$^{-}$, C$_4$$^{-}$, C$_5$$^{-}$, CO$^{+}$, CO$_2$$^{+}$, CO$_2$$^{-}$, CN$^{+}$, CN$^{-}$, N$^{+}$, N$_2$$^{+}$, N$_3$$^{+}$, N$_2$$^{+}$, N$_2$, N$_3$, N$^3$-N, NO$^+$, NO$^-$, NO$_2$, NO$_2$, N$_2$O$^+$, N$_2$O$^-$, O$^+$, O$^-$, O$_2$, O$_2$-, O$_3$, O$_3$, Cu$^+$, Cu$^-$, Cu$^{3+}$, Cu$^{+}$, Cu$^{2+}$, Cu$^{-}$ as well as electrons, are taken into account. The corresponding basic data were compiled from NASA Glenn thermodynamic database [34], NIST-JANAF thermo-chemical tables [35], and Third Millennium thermo-chemical database [36]. All mixing ratios of mixtures in the whole paper are given in molar proportion.

As an example, Figure 1 shows the compositions of CO$_2$-N$_2$ (mixing ratio 7:3) mixtures at ambient pressure, compared with the compositions contaminated by 10% Cu as shown in Figure 2. At temperatures below 5000 K, copper mainly exists in the form of Cu, Cu$_2$, and CuO, showing little influence on the fractions of oxygen and oxides (e.g. O, O$_2$, CO, CO$_2$, and NO) because the concentration of CuO is very low (<0.3%). However, at temperatures above 5000 K, due to its lower ionization energy (Cu 7.73 eV < C 11.26 eV < O 13.62 eV < N 14.53 eV [37]), copper causes the mixtures to ionize at a lower temperature. What’s more, as illustrated in Figure 3, the number density of electrons increases with copper proportion going up.

After obtaining the compositions, the thermodynamic properties (including mass density, specific enthalpy, and specific heat at constant pressure) can be determined directly according to their definitions. The corresponding formulas were given in our previous publication [14].

The effect of copper concentration on mass density and specific heat of CO$_2$-N$_2$ (mixing ratio 7:3) mixtures at ambient pressure is described in Figures 4 and 5, respectively. Obviously, with the increase of copper content, the values of mass density go up in the whole temperature range, whereas specific enthalpy drops down except at temperatures below 4500 K. This is because copper has larger molar mass (63.5 g mol$^{-1}$) than CO$_2$ (44.0 g mol$^{-1}$) and N$_2$ (28.0 g mol$^{-1}$). In addition to the difference of molar mass, more atoms (e.g. Cu) and less molecules (e.g. CO$_2$ and N$_2$) result in smaller volume because of the contribution of dissociation products of molecules, also reducing mass density. For specific enthalpy, considering that the molar enthalpies of the species composed of Cu are different from those composed of C, N, and O, the balance between molar enthalpy and mass density leads to the contrary observation below and above 4500 K.
Fig. 3. Electron number density of CO$_2$-N$_2$ (mixing ratio 7:3) mixtures contaminated by different proportions of Cu (molar proportion) at 1 bar.

Fig. 4. Mass density of CO$_2$-N$_2$ (mixing ratio 7:3) mixtures contaminated by different proportions of Cu (molar proportion) at 1 bar.

The influence of copper on specific heat was also investigated for CO$_2$-N$_2$ (mixing ratio 7:3) mixtures at 1 bar in Figure 6. As can be seen, the CO$_2$-N$_2$ mixtures with a low or medium copper concentration ($\leq$50%) have three peaks in specific heat at around 3300 K, 7200 K, and 15000 K, which correspond to the decompositions of O$_2$ and CO and ionization of N respectively. When more copper is mixed, these three peaks will disappear gradually, and instead, two new peaks at around 9800 K and 20000 K will be observed, which correspond to the first and second ionizations of copper. In general, copper reduces specific heat in the whole temperature range because of the increase of mass density (as shown above) as well as its smaller heat capacity than that of CO$_2$ and N$_2$.

3 Effects of copper on transport coefficients

The transport coefficients (including electrical conductivity, viscosity, and thermal conductivity) of a gas plasma are always useful when determining whether it can be applied in a certain industrial application. For example, in high-voltage circuit breakers (HVCB), a successful interruption requires that the arc quenching gas must have high thermal conductivity to remove energy quickly from itself [38]. The calculation of transport coefficients was based on the Chapman-Enskog method and was detailed in our previous work [17,33]. Among all the steps, the collision integrals which describe the interactions between each particles in mixtures, are the key pre-calculation [39]. They are also essential to determining combined diffusion coefficients in Section 4.

3.1 Collision integrals

In order to determine collision integrals, four kinds of interactions, namely neutral-neutral, neutral-ion, neutral-electron, and charged-charged interactions, should be considered. For charged-charged interactions, the Coulomb
potential was adopted [40]. Following our previous work [14,17], the Debye length is calculated without consideration of ions. For neutral-electron interactions, the transport cross sections of C [33], C2 [33], C3 [33], CO [33], CO2 [33], O [33], O2 [33], N [20], N2 [20], NO [20], and Cu [17] interacting with electrons were integrated to obtain collision integrals [15]. For the collisions e-Cu2, e-CuO, e-CN, and e-NO2, following the work of Cressault et al. [20], the polarizability potential was used to calculate collision integrals. The other neutral species were neglected because of their low concentration when existing with electrons. For neutral-neutral and neutral-ion interactions, the Lennard-Jones like phenomenological model potential [4,41] was preferred rather than the classical potential, for the reason presented in our previous work [17].

Figures 7 and 8 present collision integrals $\Omega(1,1)$ and $\Omega(2,2)$ for C-C, N-N, O-O, and Cu-Cu interactions, showing the much larger values of Cu-Cu than the others, which predictably can affect the transport coefficients. In Section 3.2, these collision integrals will help to explain the behaviours of transport coefficients.

3.2 Electrical conductivity, viscosity, and thermal conductivity

Using the above collision integrals, the transport coefficients of CO2-N2-Cu mixtures were calculated based on the work of Deveto [42] and Bulter et al. [43], which was validated in our previous publication [17]: electrical conductivity was calculated to a third-order approximation, neglecting the contribution of ions; viscosity was calculated to a first-order approximation without contribution of electrons; thermal conductivity was calculated as the sum of three components, namely the translational thermal conductivity, internal thermal conductivity, and reaction thermal conductivity. The results are illustrated in Figures 9–11 for electrical conductivity, viscosity, and thermal conductivity respectively. The copper proportion ranges from 1% to 99% and system pressure is constantly 1 bar.

As seen in Figure 9, the temperature range can be fragmented into three parts for electrical conductivity: 2000–4500 K, 4500–15 800 K, and 15 800–30 000 K. Below 4500 K, due to the very weak ionization as shown in Figure 3 for electron number density, electrical conductivity of mixtures is almost zero and therefore independent of copper content. From 4500 to 15 800 K, with the increase of copper and gradual enhancement of ionization, electrical conductivity is raised because copper is easier to ionize than the other species in mixtures. However, at temperatures above 15 800 K, electrical conductivity drops dramatically despite the increase of copper. This can be explained by charged-charged interactions in this temperature range. According to the work of Deveto [42], electrical conductivity is inversely proportional to collision integrals in a first approximation. Although copper
viscosity of CO$_2$–N$_2$ (mixing ratio 7:3) mixtures contaminated by different proportions of Cu (molar proportion) at 1 bar.

At 1 bar. Results in more electrons, more ions are generated simultaneously. As a result, large collision integrals between charged particles cause electrical conductivity to decrease with copper proportion [17].

Compared with electrical conductivity, the effect of copper on viscosity is more clear. As observed in Figure 10, increasing copper diminishes dramatically the viscosity of CO$_2$–N$_2$–Cu mixtures, especially from 7000 to 16 000 K. At the beginning of this temperature range (before viscosity peaks the value), atoms instead of molecules play an import role in mixtures. Considering that viscosity is in inverse proportion to collision integral [15], the large collision integrals of Cu–Cu interaction (as shown in Figs. 7 and 8) are responsible for such significant decrease of viscosity. More copper leads to more ions and therefore lower viscosity. Besides, as discussed before, copper has larger molar mass than CO$_2$ and N$_2$, which also contributes to the decrease of viscosity.

Influence of copper on thermal conductivity of CO$_2$–N$_2$ mixtures was also investigated. It is found that copper reduces thermal conductivity significantly both in the low and high temperature ranges. At low temperatures, the reaction component dominates the total thermal conductivity. The increase of copper and thus decrease of CO$_2$ and N$_2$ lead to the drop of reaction thermal conductivity because of the decomposition of molecules. At high temperatures, the translation of electrons plays the leading role in the total thermal conductivity. However, although more electrons are generated through ionization with more copper, the stronger interactions and thus larger collision integrals between charged particles result in the decrease of thermal conductivity. What’s more, similar to specific heat as shown in Figure 6, the three peaks can be observed at 3300 K, 7200 K, and 15 000 K respectively in Figure 11 for thermal conductivity of CO$_2$–N$_2$ mixtures with small fraction of copper. These three peaks can also be attributed to the dissociation of O$_2$ and CO and ionization of N respectively. When increasing copper proportion, due to the gradual reduction of O$_2$, CO, and N, the peaks of thermal conductivity will gradually diminish and vanish eventually.

4 Effects of copper on four kinds of combined diffusion coefficients

Diffusion coefficients are the essential data in describing the diffusion phenomena in thermal plasmas. To completely model the diffusion in a mixture composed of $q$ species, a total of $q(q-1)/2$ ordinary diffusion coefficients $D_{ij}$, $q-1$ thermal diffusion coefficients $D_T^i$, and $q$ mass conservation equations have to be calculated and solved [18]. To simplify this treatment, Murphy [24–27] proposed the combined diffusion coefficients, namely the combined ordinary diffusion coefficient $D_{AB}^x$, combined electric field diffusion coefficient $D_{AB}^E$, combined temperature diffusion coefficient $D_{AB}^T$, and combined pressure diffusion coefficient $D_{AB}^P$, which describe the diffusion due to composition gradients, applied electric fields, temperature gradients, and pressure gradients respectively. The calculation of combined diffusion coefficients are based on the collision integrals presented in Section 3.1, and was detailed comprehensively in our previous publication [18]. Murphy’s combined diffusion theory requires that the gases do not react with each other [44], which cannot be satisfied strictly in this work. Therefore, following the previous work [18], we choose gas A to consist of the species derived from CO$_2$ and N$_2$, and gas B the species derived from Cu as well as CuO. The results for these four kinds of diffusion coefficients of CO$_2$–N$_2$ (mixing ratio 7:3) mixtures are presented in Figures 12–15. The effects of copper on the coefficients are discussed here.

![Fig. 10. Viscosity of CO$_2$–N$_2$ (mixing ratio 7:3) mixtures contaminated by different proportions of Cu (molar proportion) at 1 bar.](image)

![Fig. 11. Thermal conductivity of CO$_2$–N$_2$ (mixing ratio 7:3) mixtures contaminated by different proportions of Cu (molar proportion) at 1 bar.](image)
Fig. 12. Combined ordinary diffusion coefficient $D_{AB}^x$ of CO$_2$-N$_2$ (mixing ratio 7:3) mixtures contaminated by different proportions of Cu (molar proportion) at 1 bar.

At low temperatures, the mean free path of neutral particles play a major role in $D_{AB}^x$ [19,20,24]. The contamination of copper changes the mean free path so weakly that $D_{AB}^x$ is almost independent of copper concentration, as observed in Figure 12. However, at high temperatures, copper raises $D_{AB}^x$ dramatically. The turning points are found to be at the round of the peaks of $D_{AB}^x$, and furthermore, the temperatures corresponding to the peaks are shifted to higher values when more copper is mixed. As discussed in the previous work [18], $D_{AB}^x$ is mainly affected by the Coulomb interactions between charged particles at high temperatures. According to standard chemical equilibrium considerations, copper with higher concentration ionizes at higher temperature, which means that the Coulomb interaction becomes important at higher temperature, causing the shift of the peak in $D_{AB}^x$.

Similar to the discussion of electrical conductivity in Section 3.2, the temperatures in Figure 13 for $D_{AB}^E$ can also be divided into three ranges: 2000–4000 K, 4000–16500 K, and 16500–30000 K. Below 4000 K, due to the negligible effect of ionization, $D_{AB}^E$ is approximately zero, whatever the proportion of copper. Above 4000 K, because of the electric field acting on the ionized mixtures, the sign of $D_{AB}^E$ reverses at nearly 16500 K. In these two temperature ranges, the influence of copper is not monotonous but changes with copper concentration. This can be explained by the balance of dependences of $D_{AB}^E$ on the mass and the ionization degree of mixtures, because $D_{AB}^E$ decreases with mass but increases with ionization degree [18].

As for $D_{AB}^T$ and $D_{AB}^P$, copper also exerts strong influence on them; generally, if the proportion is not extremely high (<90%), copper raises both the values of $D_{AB}^T$ and $D_{AB}^P$ because of the impact of its large mass. However, as illustrated in Figures 14 and 15, the further increase of copper (>90%) starts to reduce the values of $D_{AB}^T$ and $D_{AB}^P$. This is because copper leads to the increase of mass density, as shown in Figure 4, considering that $D_{AB}^T$ and

5 Conclusions

In the present work, the equilibrium compositions, thermodynamic properties (including mass density, specific enthalpy, and specific heat), transport coefficients (including electrical conductivity, viscosity, and thermal conductivity), and four kinds of combined diffusion coefficients due to composition gradients, applied electric fields, temperature gradients, and pressure gradients respectively, were calculated for CO$_2$-N$_2$ (mixing ratio 7:3) plasma contaminated by copper vapour at ambient pressure. The effects of copper on such properties are discussed. The following conclusions can be drawn:

1. Copper causes the mixtures to ionize at a lower temperature and leads the number density of electrons to increase.
(2) With the increase of copper, mass density and specific heat of the mixtures increase and decrease respectively in the whole temperature range, whereas specific enthalpy goes up and down below and above 4500 K respectively.

(3) With the increase of copper, viscosity and thermal conductivity are both reduced while the effect of copper on electrical conductivity varies in three different temperature ranges.

(4) With the increase of copper, $D_{AB}$ is raised sharply after it reaches the peak value; $D_{AB}^E$ shows the similar observation as electrical conductivity and should be considered in three temperature ranges; $D_{AB}$ and $D_{AB}^E$ are also raised when proportion of copper is below 90% in general and reduced otherwise.

(5) Copper also changes the number and position of peaks both in specific heat and thermal conductivity.

(6) The better ionization ability and larger molar mass of copper and larger collision integrals related to copper, are always responsible for its significant influences on the thermophysical properties of CO$_2$-N$_2$-Cu mixtures.

Author contribution statement

All the authors were involved in this work and contributed equally to the paper.

This work was supported by National Key Basic Research Program ("973" Program) of China (No. 2015CB251001), National Natural Science Foundation of China (Nos. 51407136 and 51521065), Fok Ying Tong Education Foundation (No. 141658). This work was also supported by the program of China Scholarship Council (CSC) for joint-Ph.D. students (No. 201506280131).

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