Kinetic study of gas heating in low pressure CO_2 plasmas

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Carbon dioxide (CO_2) is an important component of our atmosphere and a dominant constituent in the atmospheres of Venus and Mars. Atmospheric CO_2 emission need to be net-zero by 2050 to reduce global warming[1], while sustained exploration on Mars and Venus requires in-situ utilization of CO_2 resources[2]. One promising and environmentally friendly solution is to convert CO_2 into high value-added chemical products by plasma technology, due to its abundant reactive species and high chemical activity.

In recent years, numerous works have been published for CO_2 dissociation in plasma by means of experimental diagnostics, modelling, or a combination of the two. Species density, O atom loss frequency, reduced electric field, and vibrational and gas temperatures were measured in situ in a CO_2 glow discharge at low pressure[3]. These measurements provide an ideal set of constraints for validating CO_2 plasma kinetic models. A detailed self-consistent kinetic model for CO_2 plasma at low pressure was recently developed and validated[4]. However, the gas thermal balance equation was not included in the model and the gas temperature was given as an input parameter. In this work, the reaction mechanism previously established[4] is used to study the gas heating in continuous glow discharges. To this purpose, the gas thermal balance equation is added to the model to calculate the average gas temperature, assuming a gas discharge under isobaric conditions and that the heat conduction is the dominant convective cooling mechanism, and following the procedure proposed by Dias et al[5].

The calculations in this work are performed with the LisbOn KInetics (LoKI) 0D simulation tool[6], for CO_2 DC discharges at gas pressures of 1-5 Torr and currents of 10-50 mA. The electron collision cross sections and chemical reaction setup are taken from our previous work[7, 8]. In particular, two electronically excited states of CO_2 are considered, with the cross sections adopted from Phelps[9]. The contribution to gas heating adds the relaxation of CO_2 electronically excited states[8] to the traditional assumption for vibrational energy exchanges, exothermic chemical reactions, electron elastic collisions and deactivation/recombination at the wall.

Figure 1 shows the simulation results and measurements of the gas temperature as a function of the gas pressure, for various discharge currents. The gas temperature increases approximately linearly with gas pressure (1-5 Torr) and discharge current (10-50 mA). Taking the relaxation of the CO_2 electronically excited state into account for gas heating, the simulation results of the gas temperature are improved by 30 K to 50 K compared to the case where gas heating due to the quenching of electronically excited states is neglected. To further compare the simulation results and the measurements, the gas temperature is plotted against discharge power per unit length ($E*I$) in Fig. 1(b). It can be seen that the gas temperature is nearly proportional to $E*I$, and that the simulation results considering the CO_2 electronically excited state show a good agreement with the measurements.

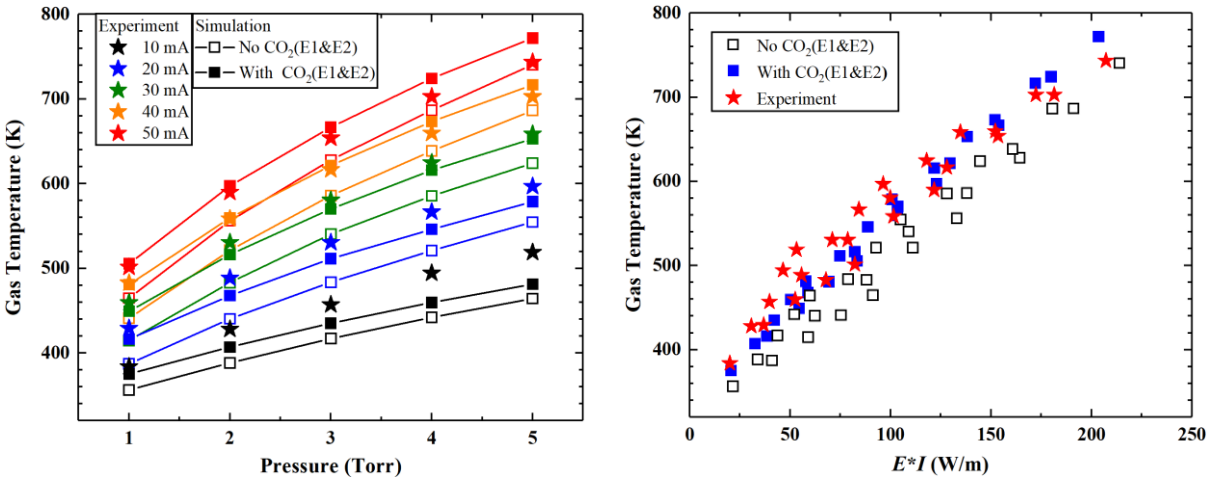

Fig. 1: Comparison between simulation and measurement results of the gas temperature against (a) the gas pressure, for discharge currents of 10-50 mA, and (b) the discharge power per unit length (*E*I*). Experimental data from [3].

It is worth noting that Naidis and Babaeva[10] have calculated the gas temperature for the conditions shown here, simply assuming that all the input energy, besides that transferred to the walls or emitted spontaneously by molecules at excited levels of asymmetric vibrational mode and that spent to dissociation through electronically excited states, goes to gas heating. Their calculations led to a very good agreement with experiment, but lack information on the detailed energy transfer pathways. The kinetic model established here provides such information. The results indicate a role of the CO_2 electronically excited states (in addition to vibrational relaxation) to the gas heating, in line with previous results obtained in pulsed discharges[8].

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