Critical assessment of reaction mechanisms using the LisbOn KInetics tool suit

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This work uses the LisbOn KInetics (LoKI) tool suit to perform a critical assessment and correction of the collisional, radiative and transport mechanisms and data describing the kinetics of several gas/plasma systems. LoKI comprises two modules: a Boltzmann solver, LoKI-B (to become open-source), and a chemistry solver, LoKI-C. Both modules can run as standalone tools or coupled in a self-consistent manner. LoKI handles simulations in any atomic / molecular gas mixture, considering collisions with any target state (electronic, vibrational and rotational), specified in the reaction mechanism adopted. As output, the tool provides the electron energy distribution function and the corresponding electron macroscopic parameters, along with the densities of species and the corresponding creation / destruction rates.

1. Introduction

Predictive tools for non-equilibrium low-temperature plasmas (LTPs) should properly describe the kinetics of both the electrons and the heavy-species, the former responsible for inducing plasma reactivity and the latter providing the pathways for reaction mechanisms. Here, we focus on plasmas produced in N_2-O_2 gaseous mixtures, aiming to deliver a KInetic Testbed for PLASMa Environmental and Biological Applications (KIT-PLASMEBA) [1], comprising the development of the LisbOn KInetics (LoKI) simulation tool, and the critical assessment of collisional-radiative data embedded in state-of-the-art kinetic schemes (KITs) for various gases and gas mixtures.

2. The LisbOn KInetics simulation tool

LoKI is a user-friendly, scalable and upgradable tool suit, developed under MATLAB® with an object-oriented design. It comprises two modules, a Boltzman solver, LoKI-B (the subject of a companion contribution [2]), and a chemistry solver, LoKI-C, that can run self-consistently coupled or as standalone tools. In Fig. 1 we can see the workflow when one or both modules are activated.

LoKI-B (to become open-source [3]) solves a time and space independent form of the two-term electron Boltzmann equation (EBE), using electron-scattering cross sections that can be downloaded from the LXCat open-access website [4]. Recently, the original capabilities of LoKI-B have been extended to obtain the time-dependent solution of the EBE for a pulsed electric field [2].

LoKI-C gives the solution to the system of zero-dimensional (volume average) rate balance equations for the heavy species (charged and neutral) present in the plasma, receiving as input data the KIT(s) for the gas/plasma system under study, and using several modules to describe the mechanisms (collisional, radiative and transport) controlling the creation / destruction of species.

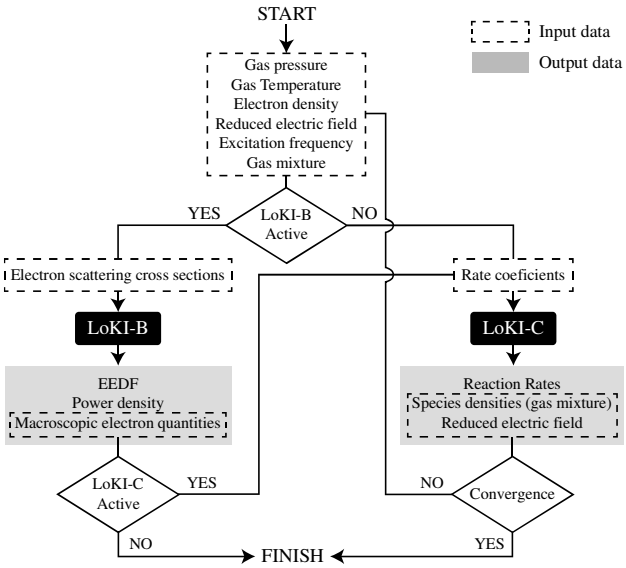

Fig. 1. Workflow of LoKI, running the Boltzmann and the Chemistry solvers in a coupled way or as standalone tools.

LoKI handles simulations in any atomic / molecular gas mixture, considering collisions with any target state (electronic, vibrational and rotational), specified in the reaction mechanism adopted. As output, the tool provides the electron energy distribution function and the corresponding electron macroscopic parameters (if LoKI-B is activated), along with the densities of species and the

corresponding creation / destruction rates. For stationary discharges, when both modules are activated, the reduced maintenance electric field (or an equivalent parameter, such as the electron temperature) is self-consistently calculated as an eigenvalue solution to the problem, under the assumption of quasi-neutrality [5,6]. Typical run times are ~500 s, for calculations done in a laptop with a single processor at ~3.5 GHz.

3. Critical assessment of the KIT for nitrogen

Verification and validation procedures are mandatory to ensure the quality of the tool and the results it provides. The validation roadmap includes the critical assessment of the collisional, radiative and transport mechanisms and data describing the kinetics of a gas/plasma system. Within this analysis, we are retrieving experimental data originally used in the validation of early model-versions, to evaluate and improve the current quality of model predictions.

Here, we show the results of a set of simulations for pure nitrogen DC discharges, obtained using the reaction mechanism presented in [6,7] with the full LoKI tool-suit. The kinetic scheme considers the vibrationally excited levels of ground-state molecular nitrogen N_2(X,v=0-59); the electronically excited states N_2(A, B, C, a, a', w); the atomic states N(^{4}S, ^{2}D, ^{2}P); and the positive ions N^+ , N_2^+(X, B), N_3^+, N_4^+. The simulations were performed for an infinitely long tube of radius 1 cm, gas pressures and temperatures in the ranges p = 0.5-2 Torr and T_g ~ 400-700 K, respectively, and discharge currents I = 5-100 mA.

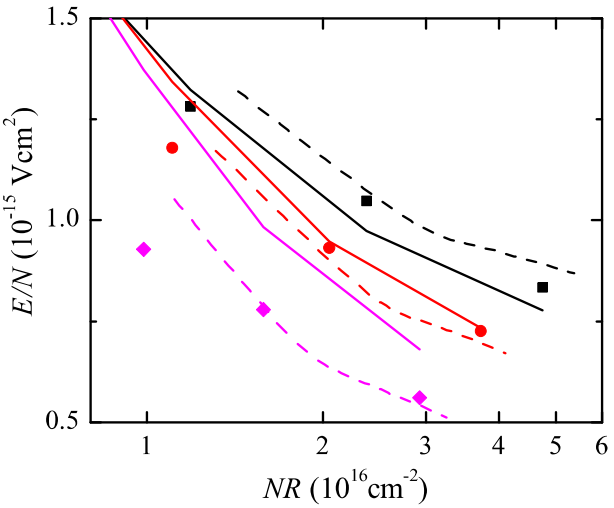

Fig. 2. Characteristics of *E/N* versus *NR* calculated (solid lines – using LoKI; dashed lines – from [8]) and measured [9] (points) at the following discharge currents (in mA): 10 (black), 30 (red) and 100 (magenta).

Fig. 2 shows, for various currents, the discharge characteristics, corresponding to the plot of the maintenance reduced electric field, *E/N*, versus the product of the gas density, *N*, and the tube radius, *R*. The figure shows calculations obtained using either LoKI or a previous in-house code and model [8]. We observe a fair agreement between calculations and measurements for the higher *NR* values, which differs according to the model/code adopted. These results evidence the importance of pursuing the critical analysis of mechanisms and data. Yet, the first calculations obtained with LoKI and the updated reaction mechanism of [7] already give good predictions for the populations of molecular excited species, such as N_2(A) and N_2(B), as a function of the discharge current, as represented in Fig. 3.

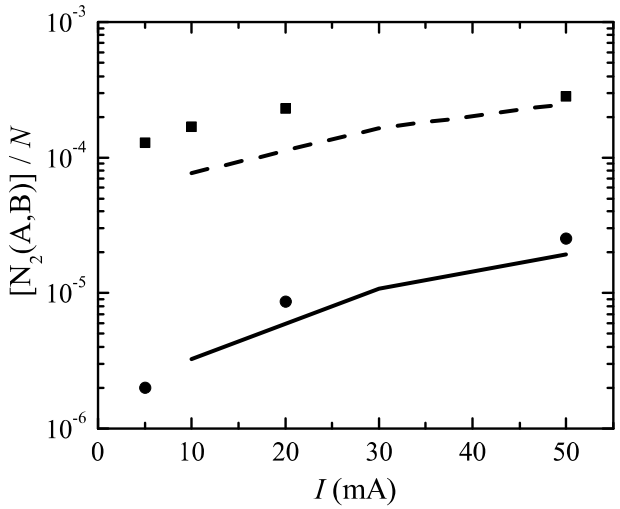

Fig. 3. Fractional concentration of N_2(A) (dashed lines and squares) and N_2(B) (solid lines and circles), vs I at p = 1 Torr, calculated using LoKI (lines) and measured [9] (points).

Work is in progress to update the kinetic schemes for nitrogen (and other gases and gas mixtures), in order to improve the predictions of the simulations.

Acknowledgments

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