Genetic algorithm-based investigations on the key chemical reaction pathways in a helium-air cold atmospheric plasma jet

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Abstract: Spatiotemporal evolutions of the chemically reactive species in a cold atmospheric plasma (CAP) system are important for both fundamental studies and applications. Since many possible chemical reaction pathways may be involved in a CAP system, it is indispensable to investigate the relative importance among the numerous possible reactions. In this study, a numerical method based on the genetic algorithm is proposed to determine the key chemical reactions in a helium-air CAP jet with a high calculation efficiency.

Keywords: Cold atmospheric plasma, chemical reactions, genetic algorithm coupled with dynamic programming

1. Introduction

Cold atmospheric plasmas (CAPs) which contain multiple chemically reactive species have promising application prospects in numerous fields such as bio-medicine, agriculture, synthesis or modification of advanced micro- / nano-scale sized materials, in-door atmosphere purification, etc. The spatiotemporal evolutions of the chemically reactive species in a CAP system is of great importance not only for understanding the non-equilibrium characteristics of plasmas, but also for achieving excellent plasma processing results in actual applications. Except for experimental diagnostics, numerical simulations considering complex chemical kinetics and reaction dynamics have been conducted to investigate the non-equilibrium characteristics of the CAPs. However, on one hand, taking a great number of chemical reactions into consideration will cause great complexity and instability for the numerical simulations; while on the other hand, we have noticed that the variation trends of the same species under the same or similar operating conditions may be quite different provided in different studies because the chemical reaction models were different [1, 2], as shown in Fig.1. Therefore, it is of great importance to determine the key chemical reaction pathways among numerous possible reactions.

For the studies aiming to understand the complex plasma chemical dynamics, a global model has been employed widely. As a volume-averaged model, it is assumed that the plasma is homogeneous in space focusing on the complex chemical kinetics. However, it is usually time-consuming due to the involvement of numerous possible chemical reactions with many possible chemically reactive species in a global model. Therefore, a numerical method based on the genetic algorithm coupled with a dynamic programming is proposed in this study to determine the key chemical reaction pathways in a CAP system with a high calculation efficiency.

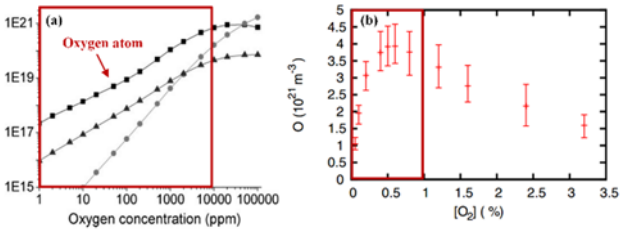

Fig. 1. Different variation trends of the concentrations of oxygen atoms provided in different studies [1,2].

2. Theoretical Method

In this study, with a helium-air CAP jet driven by a radio-frequency (RF) power supply (Fig. 2) as a model system, selections on the dominated chemical reaction pathways are conducted based on a 0-dimensional (0-D) numerical simulation. A coaxial-type plasma generator with a water-cooled bare-metallic electrode configuration is employed to produce the glow discharge plasmas, which is the so-called RF-APGD plasma hereafter, with a (97.5%) helium–nitrogen gas mixture as the plasma working gas. The inner electrode works as the powered electrode connecting to the 13.56 MHz RF power supply, while the outer electrode is grounded.

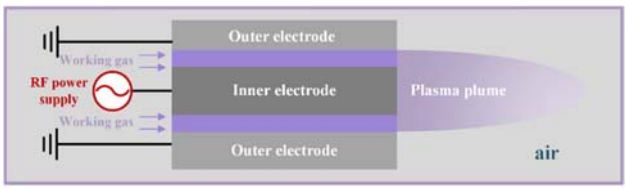

Fig. 2. Schematic of the RF-APGD plasma generator.

Since we focus on the chemical kinetics in the plasma jet region, the time-averaged one-dimensional (1-D) modelling results in the discharge region [3] are used as the initial condition in this study. Beginning with a comprehensive chemical reaction model involving 28

species and 135 reactions, the 0-D modelling is conducted focusing on the temporal evolutions of the chemically reactive species concentrations and the electron energy. The governing equations to be solved simultaneously are listed as follows.

(i) Species mass conservation equation:

$$\frac{dn_i}{dt} = S_i \left(i = 1, \ \ldots, \ 28\right) \quad (1)$$

where n_i and S_i are the number density and homogeneous production/destruction rate of species i, respectively.

(ii) Electron energy conservation equation

$$\frac{d}{dt}\left(\frac{3}{2} n_e k_B T_e\right) = -\sum_j \Delta E_j^e K_j - 3\frac{m_e}{m_{He}} n_e \bar{\nu}_e k_B \left(T_e - T_g\right) \quad (2)$$

where n_e, T_e and m_e are the number density, temperature, and mass of electrons, respectively, while k_B, ΔE_j^e, K_j, $\bar{\nu}_e$ and T_g are the Boltzmann's constant, energy loss during the inelastic collision process, the corresponding reaction rate, the momentum transfer rate between electrons and background particles, and the gas temperature, respectively.

Since not all the possible chemical reactions considered in the numerical models affect the chemical reaction kinetics in plasmas equally [4], the aim of the chemical model simplification is to find a set of reactions with as less number as possible (reduced chemical reaction model, RCRM) to substitute all the possible reactions (comprehensive chemical reaction model, CCRM) with a relative discrepancy lower than a certain critical value. To achieve this target, the influences of each reaction on the chemical reaction kinetics should be evaluated, which results in a great amount of work. Therefore, the genetic algorithm coupled with a dynamic programming is proposed in the present paper to solve this problem.

In the genetic algorithm, the chromosome is defined as a vector $\boldsymbol{k} = (k_1, k_2, k_3, ..., k_{135})$ and the value of k_j (j = 1, 2, 3, …, 135) is either the rate coefficient of the jth reaction or zero (zero means that the corresponding reaction is out of consideration). Therefore, every chromosome corresponds to a possible selection of the reduced chemical reaction model. Our goal is to find a chromosome that not only can substitute the original CCRM with an acceptable relative discrepancy, but also contains the least nonzero elements.

As shown in Fig.3, first, the chromosomes are initialized and several couples of parent chromosomes are selected to obtain the child chromosomes through crossover, reproduction, and mutation. Every child chromosome will lead to a set of modelling results through the global model. Then, the fitness (δ_{max}) can be calculated, which is defined as the maximum relative discrepancy between the original CCRM and the RCRM corresponding to the child chromosome. Finally, we can judge if the iteration has reached convergence, and decide to enter the next iteration or end the calculation.

However, because the genetic algorithm cannot guarantee that the finally obtained chromosome has the least nonzero elements, leading to massive times of iterations if the initial chromosomes are chosen badly. Therefore, a dynamic programming is coupled into the genetic algorithm in this study.

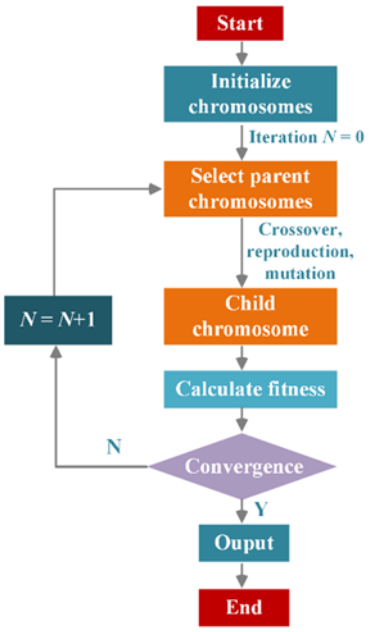

Fig. 3. Flowchart of the genetic algorithm.

If we define the status-k in our algorithm as a serial of chromosomes which includes k nonzero elements, the main idea of the dynamic programming is to find the optimal solution according to the previously acquired solutions. As illustrated in Fig. 4, starting from k = 1, if the fitness (δ_{max}) is lower than the pre-set critical value (δ_c) in a certain status-k (e.g., 10% used in this study), this means that the corresponding status-k is the optimized solution; otherwise, we will move to the next status with (k+1) to solve the 0-D numerical model using the status-k as the newly updated chromosomes. The calculation will end when the criterion $\delta_{max} < \delta_c$ is satisfied with $k = l$. Thus, it is seen from the flowchart of Fig. 4 that the finally obtained optimized chromosomes, status-l, contains the least nonzero elements.

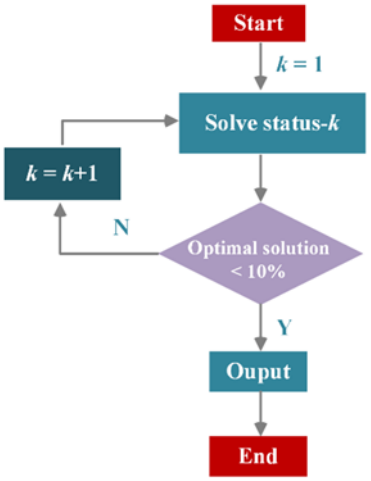

Fig. 4. Flowchart of the dynamic programming.

3. Results and Discussions

Starting from a certain initial condition including 135 reactions and based on the genetic algorithm coupled with the dynamic programming, a RCCM, including much fewer chemical reactions than the original CCRM with 135 reaction pathways, is obtained with δ_c = 10%. In addition, further validations are also conducted with different initial conditions. For example, when the initial electron number densities vary from 10^{14} to 10^{16} m^{-3}, the modelling results show that the corresponding values of δ_{max} are all less than 10%. This indicates that the obtained RCCM can reasonably substitute their corresponding CCRM with a high calculation efficiency.

4. Conclusions

In this study, a numerical method based on the genetic algorithm coupled with the dynamic programming is developed to simplify the chemical reaction pathways with a helium-air CAP jet as a model system for improving the calculation efficiency, while simultaneously with enough calculation accuracies. The modelling results indicate that there exist much fewer chemical reaction pathways, comparing with the original 135 reactions, which overwhelmingly affect the evolutions of the species number densities and electron energies in the helium-air CAP jet. This also provides a high-efficient general method for simplifying the chemical reactions in different plasma systems.

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References

[1] D.-X. Liu, M.-Z. Rong, X.-H. Wang, F. Iza, M. G. Kong, P. Bruggeman, Plasma Processes and Polymers, **7**, 846 (2010).
[2] M. M. Turner, Plasma Sources Science and Technology, **24**, 035027 (2015).
[3] X.-F. Zhang, Z.-B. Wang, Q.-Y. Nie, H.-P. Li, C.-Y. Bao, Applied Thermal Engineering, **72**, 82 (2014).
[4] M. M. Turner, Plasma Sources Science and Technology, **25**, 015003 (2015).