

CHARACTERISTICS OF ARGON AND HELIUM PLASMAS CREATED BY MICROWAVE DISCHARGE AT ATMOSPHERIC PRESSURE

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Abstract: A review of recent developments in the field of microwave plasmas created in noble gases at atmospheric pressure is presented. Several possible designs of discharge configuration are presented and the evolution of plasma jet is explained. The contraction and filamentation of plasma is explained and illustrated. Such plasma is characterized by a high temperature of both neutral and ionized gaseous atoms, which can easily reach several 1000K. The electron temperature is often between 15000 and 30000K, and the electron density usually exceeds 10^{20} m^{-3} . At such conditions, several reactions untypical for low pressure plasmas occur. Among them, formation of dimmers is of particular interest. In some cases, the density of dimmers such as He_2^+ may exceed the density of common He^+ ions.

Značilnosti argonske in helijeve plazme ustvarjene z mikrovalovno razelektritvijo pri atmosferskem tlaku

Ključne besede: mikrovalovna plazma, atmosferski tlak

Izvilleček: V prispevku opisujemo zadnja dognanja na področju mikrovalovne plazme, ki jo ustvarimo v žlahtnih plinih pri atmosferskem tlaku. Opisujemo nekatere možne konfiguracije razelektritve in razložimo razvoj plazemskega curka znotraj razelektritve. Curek plazme se pri teh razmerah običajno zoži znotraj razelektritvene cevi, pojavijo pa se tudi plazemske niti, kar ilustriramo in razložimo v tem prispevku. Takšno plazmo odlikuje visoka temperatura nevtralnega plina in ioniziranih atomov, ki zlahka doseže več 1000K. Temperatura elektronov je pogosto med 15000 in 30000K, medtem ko njihova gostota običajno preseže vrednost 10^{20} m^{-3} . Pri takšnih razmerah opazimo nekatere reakcije, ki niso značilne za nizkotlačne plazme. Med njimi je posebej zanimiva tvorba dvoatomnih molekul žlahtnih plinov. V nekaterih primerih, kot npr. He_2^+ , lahko gostota takšnih molekul celo preseže gostoto običajnih helijevih ionov He^+ .

1. Introduction

High frequency plasmas are widely spread sources of active species which can be used in various applications /1-42/. When these sources are set up on processes working at atmospheric pressure, lower cost treatments can be achieved if efficiency is unchanged with respect to processes under low pressure. Therefore, there is a need to better understand and characterize high pressure sources to optimize the treatments /43-59/.

Microwave plasmas in CW mode operate at high treatment temperatures, namely a few thousands Kelvin. Several sources are widely studied like surface wave excited plasmas /60-64/, waveguide-based microwave torches, like the "Torche à Injection Axiale" /65-72/ and resonant cavities /73-76/. In Fig. 1, three sources are presented showing possible designs for atmospheric pressure microwave plasmas. The plasma is either created in open air, confined without wall contact or guided by a fused silica tube where the electric field propagates. Some of these sources cannot operate at high power since fused silica which is commonly employed as confinement vessel melts at ~2000 K. Discharges in rare gases are mostly studied, especially in argon and helium. Beyond design and arrange-

ment of the sources, strong efforts, both theoretical and experimental, were performed recently to better understand the physical processes that govern these plasmas. However, complementary studies are still necessary to improve our knowledge on these small-scale plasmas where huge gradients exist. Indeed, diagnostics with high spatial resolution are needed. Some basic data for kinetic processes and even for some species should be determined experimentally at high temperature. This would help develop predictive self-consistent collisional radiative models in optically thick media.

In the present paper, we review recent advances in microwave sources at atmospheric pressure. We describe the difficulties to overcome in order to progress in the understanding of these plasmas. The outline of the paper is then as follows: In section 2, a special attention is paid to recent theories available to describe contraction of some rare gas plasmas. A brief comment will also be given on filamentation. In section 3, we present the most relevant question on basic data needed in the field of atmospheric microwave sources. Finally, concluding remarks are provided in section 4.

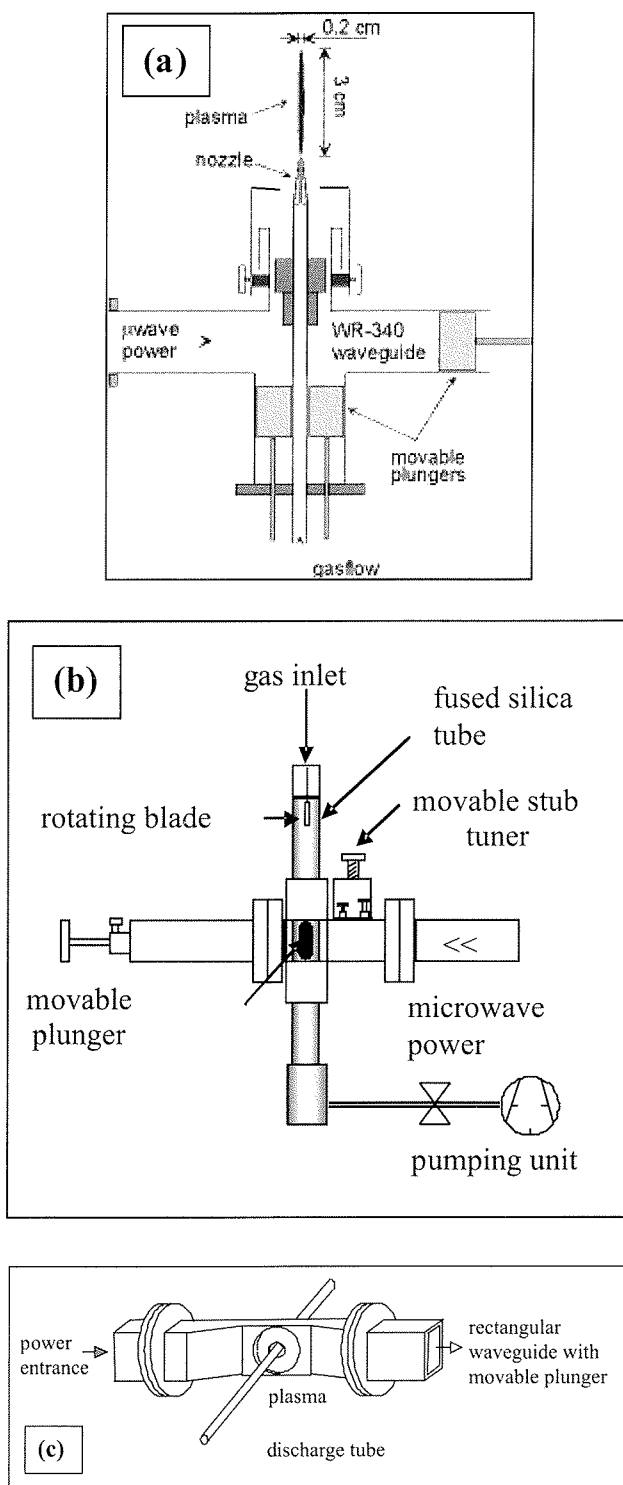


Fig. 1: Some possible designs of microwave sources operating at atmospheric pressure. (a) The "Torche à Injection Axiale" /65/, (b) resonant cavity /73/ and (c) surfaguide wave launcher /60/. The plasma can be sustained in open air, confined without wall contact or guided along a dielectric, respectively.

2. Contraction and filamentation in rare gas plasmas

Contraction can be defined as the compression of plasma into a filament located at the discharge axis. It can be observed in various gases and excitation sources, not only microwave (see Fig. 2). It is characterized by a local increase in both the electron density and the gas temperature and by a decrease in the electron temperature. In the other hand, filamentation is observed only in high frequency discharges only. One single plasma filament splits into two or more filaments of smaller diameters /77, 78/ once the electron density is sufficiently increased radially.

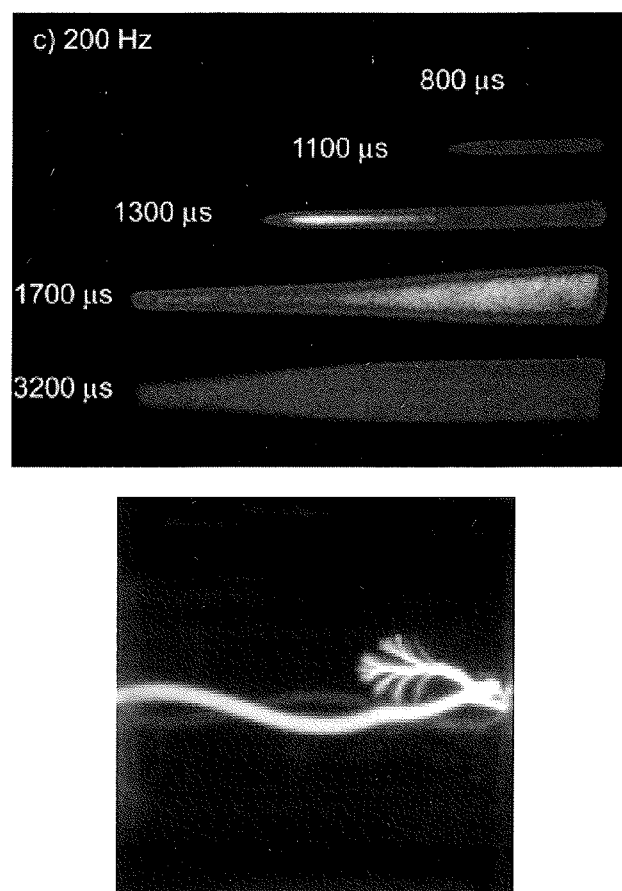


Fig. 2: Illustration of contraction (left) and filamentation (right) phenomena. Left figure is the time evolution of a neon plasma column sustained at atmospheric pressure at 200 Hz (after /78/). Right figure is an argon plasma sustained in a microwave resonant cavity at atmospheric pressure pulsed at 2kHz (duty cycle is 41%).

Recently, study of contraction has gained interest because of the potentialities of these plasmas at atmospheric pressure. Two main mechanisms relying on the same idea, the radial decrease in the ionization rate, can be evoked to explain the contraction process. The first one deals with electron-electron collisions. Energetic electrons which are depleted by inelastic collision can be heated by electron-electron collisions that tend to give a Maxwellian distribu-

tion. However, this mechanism is only possible if the electron density is high enough, usually close to the discharge axis. Then, at the edge of the plasma, the tail of electron energy distribution function is likely depleted and the ionization rate decreases radially /79-80/. The second possible explanation is nonuniform heating of the gas along the discharge radius. It creates contraction through its influence on the kinetics of dimer ions, which controls the charged-particle balance. These descriptions rely on basic data that now need to be examined in detail for rare gases. Helium and argon are considered next.

3. Basic data

A common difficulty present in available works deals with the temperature dependence of rate constant of chemical reactions over wide ranges. Indeed, depending on the power delivered to the plasma, the temperature of the gas can vary over large range of values. Moreover, due to high temperature gradients, it varies spatially. The main consequence of this change in the gas temperature is the control of the kinetics of excimers, whether they are neutral or charged. There is no measurement on the excimer density nor on their vibrational distribution above 1500 K. In Fig. 3, we give an overview of the gas temperature dependence of the rate constant for the three possible recombination processes of He_2^+ ions /81-84/. Available data are spread over a wide range of values and experimental data are all given below 600 K.

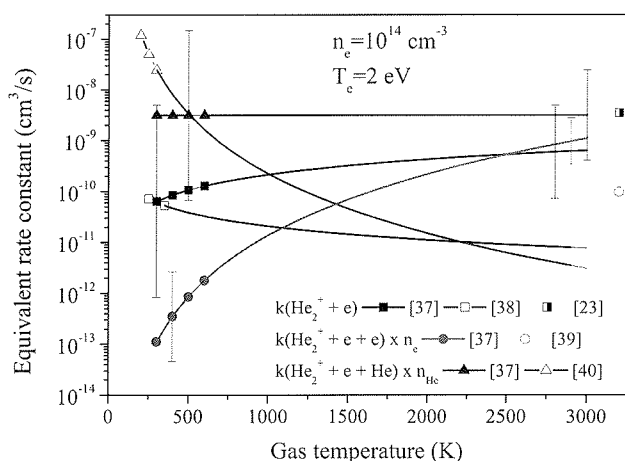
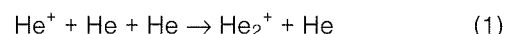


Fig. 3: Equivalent two-body rate constants for recombination processes of He_2^+ ions as a function of the gas temperature. An electron temperature of 2 eV and an electron density of 10^{14} cm^{-3} are chosen. Data deduced from experiments are by /81-/84/. Data used by /67/ is added for comparison. Extrapolations of available temperature dependences over the range [300-3200 K] are also given (solid lines). When available, the accuracy of the equivalent rate constant is reported.

Let's consider for example the case of He^+ and He_2^+ ions in helium atmospheric microwave discharges where several difficulties arise. Dimer ions are mainly produced from He^+ by a three-body collision:



with a rate constant of $1.2 \pm 0.2 \times 10^{-31} \text{ cm}^6 \text{ s}^{-1}$ at 300 K /85, 86/.

First, the temperature dependence of this process, extrapolated from data by Böhringer /85/ measured between 50 K and 350 K ($T_g^{-0.60 \pm 0.10}$) is rather different from that determined by Russel /86/ between 80 K and 300 K ($T_g^{-0.38 \pm 0.06}$). However, the two sets of experimental data differ by less than 30%. The accuracy of the measurements within this range of temperature is probably not high enough to determine correctly the exponent of the power law. The former dependence was used for example by /67/ to determine to role of dimer ions between 500 K and 3000 K.

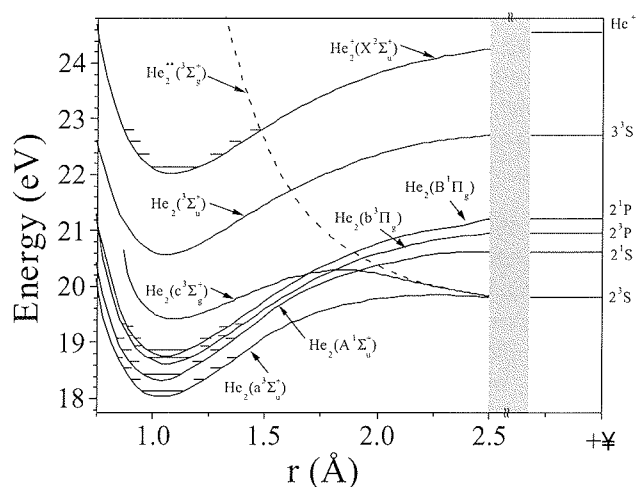
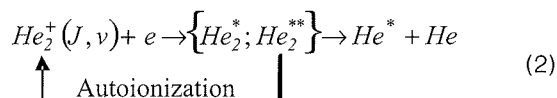


Fig. 4: Energy diagram of helium dimers. The doubly excited state He_2^{**} is an antibonding triplet state intersecting the dimer ion close to the $v=3$ vibrational level.

Second, to estimate the ratio $[\text{He}_2^+]/[\text{He}^+]$, three main processes are considered by /67/ to account for the reaction pathways possibly followed by He_2^+ : direct electron recombination, He_2^+ creation by reaction (1) and loss by the reverse process, dissociation of dimer ions by electron impact being negligible. Assuming the Saha equilibrium, the rate constant of the reverse process of reaction (1) is estimated. This choice gives a ratio $[\text{He}_2^+]/[\text{He}^+] \sim 0.55$ at 2500 K for $n_e = 10^{14} \text{ cm}^{-3}$, i.e. similar number densities for the two species. The rate constant used for direct electron recombination is $5.0 \times 10^{-9} / T_e^{0.5} \text{ cm}^3 \text{ s}^{-1}$ where T_e is the electron temperature in eV. This rate constant is given with no gas temperature dependence. However, it does depend on it as shown in recent works based on ring storage experiments. Indeed, due to the unfavourable location of the doubly excited potential energy curves (see Fig. 4), the recombination route and rate (Eq. (2)) of ions depend

both on the electron energy and the initial rovibrational distribution (/87-89/).



A competing process has to be taken into account, namely the autoionization of the intermediate Rydberg He_2^* or doubly excited He_2^{**} neutral molecule, which can fragment back into an electron and a molecular ion in the (J', v') rovibrational level.

The unusually low values of the direct recombination rate found in experimental works are at least partially explained (see /82, 90/ for details). The dependence of the rate of the direct recombination process with the electron temperature was determined to be $\propto T_e^{-0.9}$ /90/. The gas temperature dependence is not available on a large range of temperatures. However, such dependence is available for neon dimer ions /91/. Results obtained confirm a fall-off in efficiency of the direct recombination for vibrationally excited rare-gas dimer ions with increasing temperature, the contribution of autoionisation being not negligible /92/. Direct recombination should only be considered at low temperature and high electron energy.

Therefore, the two-body recombination of electrons with dimer ions should not be considered at high temperature in the estimation made by /67/. In fact, this consideration does not change significantly the predicted result. Despite ambipolar diffusion prevails at 2500 K and is much lower than direct recombination as used by /67/, the density ratio $\text{He}_2^+/\text{He}^+$ is nearly the same than that these authors found. One of the two processes, the atom assisted association or its reverse process, has rate always much higher than the loss of He_2^+ by recombination or ambipolar diffusion which are known to be slow recombination processes. If the absolute value of the density of He_2^+ depends on direct and reverse rates of atom assisted association, the density ratio $\text{He}_2^+/\text{He}^+$ does not (see Eqs. 10 and 11 p 466 in /67/). In fact, to modify the temperature dependence of the density ratio $\text{He}_2^+/\text{He}^+$, one should either increase the rate of the recombination or decrease the rate of dissociation by atom impact. A last remark deals with the role of associative ionization. Rate constants for these processes at high temperature are unknown and the reaction processes should be included in the calculation of the density ratio $\text{He}_2^+/\text{He}^+$ if they are not negligible.

Considering now the temperature dependence by Russel /86/, if the balance of association and dissociation via atom impact is in equilibrium, one should find a ratio $\text{He}_2^+/\text{He}^+ \sim 9.2$ at 2500 K for $n_e = 10^{14} \text{ cm}^{-3}$, showing that at high temperature, He_2^+ ions may possibly still prevail.

In argon, these same authors find a ratio $\text{Ar}_2^+/\text{Ar}^+ \sim 3.0 \times 10^{-3}$ at 2500 K for $n_e = 10^{15} \text{ cm}^{-3}$, a result used by /63/ more recently. This difference between helium and argon is attributed to the dissociation energy of the dimers (2.4 eV

versus 1.3 eV), which is nearly twice higher for the former and only partially compensated by the larger partition function of argon ion dimers. Here again, no experimental data is available to determine how the $\text{Ar}_2^+/\text{Ar}^+$ ratio evolves versus gas temperature.

The same kind of problem arises for excimer creation or associative ionization despite theoretical data are available in the case of this latter processes for some triplet states of helium /87/. However, only experimental measurements would confirm the validity of available estimations.

4. Conclusion

Atmospheric microwave plasmas are high-temperature non-equilibrium media where complex phenomena that are still under investigation occur. Better understanding these media requires specific measurements to determine rate constants at high temperature. Diagnostics with high spatial and temporal resolutions are needed to provide basic data that lack today to provide satisfying description of these sources of active species.

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