Experimental Studies on the Dual-Frequency, Atmospheric-Pressure, Dielectric Barrier Discharge Plasmas

Guo Li, Pei-Si Le, Sen Wang, He-Ping Li*, and Cheng-Yu Bao

Department of Engineering Physics, Tsinghua University, Beijing 100084, P. R. China

(* Corresponding author, H.-P. Li: liheping@tsinghua.edu.cn)

Abstract: In this study, the uniform, stable, argon radio-frequency (RF) dielectric-barrier discharges (DBDs) at atmospheric pressure are obtained with the aid of the atmospheric-pressure kilohertz DBDs. The experimental results show that by employing the dual-frequency plasma generator, a large volume, uniform, atmospheric argon RF DBDs can be achieved with a low RF breakdown voltage.

Keywords: Atmospheric pressure; DBD; dual-frequency; breakdown; argon

1. Introduction

In recent years, atmospheric-pressure glow discharge (APGD) plasma sources driven by a radio-frequency (RF) power supply are developed to obtain non-equilibrium gas discharge plasmas with large-area, high stability, uniformity and reactivity. In our opinion, there are two aspects which are very important for promoting the actual applications of the RF APGD plasma sources in a variety of fields, such as plasma-based sterilization/disinfection of the reusable heat-sensitive medical instruments contaminated with micro-organisms [1], gene mutation of micro-organisms [2], decontamination of chemical/biological warfare agents [3], etc. One aspect is the sustentation of the glow discharges with large area and stability, while at the same time, with high concentrations of the chemically active species and reactivity; the other aspect is the production of the glow discharges using gases with different chemical compositions, especially using the cheaper gases (e.g., nitrogen or air) at low breakdown voltages.

In the case of RF APGDs generated with a pair of bare metallic electrodes, there exist two different discharge modes – α-mode at low current densities which is sustained by volumetric ionization process and γ-mode at high current densities in which ionization by secondary electrons from the electrode surfaces is important [4, 5]. In actual applications, large discharge area/volume and high concentrations of chemically active species in the plasma discharge and/or after-glow regions are critical for improving the action efficiencies. But unfortunately, for the RF APGDs produced with bare metallic electrodes, the α mode discharges are usually associated with high stability and uniformity in large volume secured by simply restricting the input RF power below an appropriate level, which subsequently leads to lower concentrations of the chemically active species and poor reactivity [6]; while on the other hand, high discharge current densities would turn the glow discharges from α-mode to γ-mode producing massive reactive species with shrinking the large volume RF glow discharge into a constricted plasma column. Therefore, it is a key issue to expand the discharge area and stability of the APGDs simultaneously keeping the high current density and plasma reactivity. In Ref. [7], the RF dielectric-barrier discharges (DBDs) in atmospheric argon were reported with larger discharge area and high uniformity operating at a higher discharge current density compared with that of the RF APGDs using bare metallic electrodes.

For the RF APGD plasmas with bare-metallic electrodes, usually expensive helium is employed as the primary plasma working gas which leads to the relatively high capital costs of this technology in actual applications. The difficulties for obtaining the uniform glow discharges at atmosphere using bare metallic electrodes lie in the facts that: at atmospheric pressure it is difficult to ignite an argon, air or nitrogen discharge due to the very high Townsend breakdown threshold values (e.g. ~3.2-3.5×10^6 V/m for air and nitrogen, and ~2.7×10^5 V/m for argon at one atmosphere [8]); and even the discharge is ignited (e.g. argon), it tends to transfer into a filamentary arc after breakdown at one atmosphere due to the intense avalanche of electrons under relatively large electric field [9]. For avoiding the formation of the filamentary arc after gas breakdown at atmospheric pressure, the argon RF APGDs were generated between two parallel stainless-steel plates each covered with a ceramic sheet of 0.5 mm in thickness and 9.0 in relative permittivity in Ref. [7]. But in such case, the gas breakdown occurred at a peak applied voltage of 1150 V (the corresponding root-mean-square (rms) value is 813.3 V) at a fixed gap spacing of 2.0 mm, which is higher than that for the case with bare metallic electrodes, due to the addition of the two ceramic sheets between electrodes. In Ref. [10], by adding a small amount of ethanol into argon, the breakdown voltage can be reduced significantly and a pure α-mode discharge can be...
achieved more easily between two water-cooled, bare metallic electrodes possibly due to the Penning ionization effect. And in Refs. [11-13], the RF APGD plasmas using the cheaper gas (e.g., nitrogen, air, etc) as the primary plasma-working gas were generated also between two bare metallic electrodes using the gas induced discharge approach or under an intensified electric field with an appropriate discharge process or the design of the plasma generator, respectively, and sustained under lower applied voltages between electrodes.

Based on the preceding discussions, at the present time, it is still an interesting job to produce large volume, uniform and stable glow discharges at atmospheric pressure using the gases with high Townsend breakdown threshold values (e.g., argon, nitrogen, air, etc). As indicated in Refs. [9, 14], it would be helpful to form a uniform glow discharge if enough seed electrons can be obtained before the occurrence of another discharge under a low electric field. In this paper, two power supplies with frequencies of 13.56 MHz and 50 kHz are employed to generate the argon atmospheric-pressure DBD (APDBD) with a co-axial-type plasma generator, in which the kilohertz DBDs are employed to initiate the RF DBDs under a low applied RF voltage. The discharge characteristics, as well as the influences of the kilohertz DBDs, on the RF DBDs are discussed.

2. Experimental setup

A schematic diagram of the experimental setup is shown in Fig. 1, which consists of the power supply sub-system (RF power supply at 13.56 MHz and high-voltage alternating current (HVAC) power supply at 50 kHz), plasma generator, gas supply and control sub-system, electrical characteristic measurement sub-system and a digital camera used for recording the discharge images. The schematic diagram and the pictures of the coaxial-type plasma generators used for generating the argon DBDs at atmospheric pressure are shown in Figs. 1 and 2, respectively. The inner electrode (tungsten rod) is inserted into a 3.0-mm-inner-diameter glass tube which is sealed on one end as a grounded electrode. The rod) is inserted into a 3.0-mm-inner-diameter glass tube. The RF/HV AC powered electrode are segmented copper foils attached to the outer 1.0-mm-in-thickness glass tube. The plasma working gas is admitted into the annular space between the two glass tubes, ionized by the applied RF/HV AC electric fields, and flows out of the plasma generator. The geometrical differences between the Type I and II plasma generators are as follows: (i) the thickness of the inner glass tubes are 1.0 and 0.5 mm for the Type I and II plasma generators, respectively; (ii) the gap spacing between the glass surfaces are 1.0 and 2.5 mm, respectively, for the Type I and II plasma generators. Thus, the corresponding radial distances between the metal electrodes (RF/HV AC powered copper electrode and grounded tungsten electrode) are 3.0 and 4.0 mm for these two plasma generators. In this study, the HVAC powered electrode is located at the upstream of the RF powered electrode in the flow direction with 10.0 mm inter-distance between these two powered electrodes. The applied HVAC voltage is controlled by an adjustable transformer, while the RF powered electrode is connected to the power supply through a matching network. The rms values of the voltage and current connecting to the RF powered electrode are measured using a high voltage probe (Tektronix P5100) and a current probe (Tektronix TCP202), respectively, and recorded on a digital oscilloscope (Tektronix DPO4034). The discharge images are taken by a digital camera (Fujifilm FinePix S9600).

3. Experimental results and discussions

3.1 Discharge images

Typical discharge images using different plasma generators and under different operation conditions are shown in Fig. 3 with the constant argon flow rate \(Q_{Ar}=5.0\) slpm (standard liters per minute). The measured parameters, including the rms values of the discharge voltage and current connecting to the RF powered electrode, the operation conditions, as well as the exposure time of the camera, are listed in Table 1, in which Cases (b) and (f) represent the situations before the stable RF discharges occur with the existence of the HVAC DBDs. Here, we call the discharges driven by the RF and HVAC power supplies as the RF DBD and HVAC DBD, respectively, for simplicity. And the breakdown voltage is defined as the minimum applied RF voltage which can maintain a stable RF DBD after the HVAC DBD is turned off.

In this study, there are filaments exist in the HVAC DBDs, as shown in Figs. 3(a) and (e). By comparing Figs. 3(a) and (b), 3(e) and (f), it can be seen that before the
stable RF discharge occurs, the emission intensities of the plasmas in the space between the RF and HVAC powered electrodes increase with the increase of the of RF power or current. This phenomenon indicates that the upstream HVAC DBDs can gain energy from the RF power supply, probably because the electric field in the mid-region between these two electrodes is determined not only by the upstream HVAC electric potential, but also by the downstream RF potential with a short inter-distance between these two powered electrodes.

For the cases using the Type I plasma generator, after the stable RF DBD is obtained, the originally existing filaments in the HVAC DBDs still maintain in the RF discharge region and the mid-region between the RF and HVAC powered electrodes (Fig. 3(c)); and with the increase of the RF input power or current, the discharge region extends upstream and downstream still with the existence of the bright filaments, as shown in Fig. 3(d). That is to say, the RF DBD operates in a filamentary-glow co-existing mode for the cases with the Type I plasma generator.

For the cases using the Type II plasma generator, the thickness of the inner glass tube is reduced from 1.0 mm (Type I) to 0.5 mm (Type II), while at the same time, the gap spacing between electrode is increased from 1.0 mm (Type I) to 2.5 mm (Type II). By comparing the discharge images in Figs. 3(e) and (f) with those in Figs. 3(a) and (b), it can be seen that due to the change of the plasma generator geometrical dimensions, the emission intensity and the number of the filaments for the HVAC DBDs are reduced significantly. After the stable RF DBD occurs, the filaments can disappear if the HVAC power is turned off, and thus, a uniform glow mode discharge can be obtained, as shown in Fig. 3(g); then, with the increase of the RF current or power input, the volume of the glow mode discharge enlarges, leading to the increase of the plasma jet length at the downstream of the RF powered electrode and the increase of the emission intensities of the plasmas (Fig. 3(h)); and then, when the RF current is higher than 198.3 mA (the corresponding RF voltage is 579.6 V), a mode transition from the glow mode to the filamentary-glow co-existing mode occurs in the extended discharge region, as illustrated in Fig. 3(i), and the discharge becomes unstable.

The discharge phenomena presented in Fig. 3 show that the geometrical dimensions and the RF power input have pronounced influences on the discharge modes and stability.

### 3.2 Electrical characteristics

The voltage-current ($V-I$) curves of the pure argon RF discharges with the help of the upstream HVAC discharges by using the dual-frequency plasma generators are shown in Fig. 4 at $Q_{Ar}=5.0$ slpm. In this paper, after the stable RF discharge appears, i.e., the breakdown process defined in this paper occurs, the HVAC power supply is turned off. For both types of the plasma generators, the $V-I$ curves before breakdown in Fig. 4 are bee-lines because the generators can be regarded as capacitors. Due to the existence of the upstream HVAC DBDs, the RF breakdown occurs at relative low voltages, i.e., 454.7 V and 352.4 V at Point B and D for the Type I and II plasma generators, respectively. The contrast experiments show that, with other parameters being unchanged, the RF breakdown of argon cannot occur for both of the plasma generators even if the applied voltages are as high as 1046 V, which is the maximum output voltage of the RF power supply, if no help of the HVAC DBDs due to the high Townsend breakdown threshold value of argon at one atmosphere (~2.7×10⁵ V/m [8]).

In this study, there are no significant voltage drops occur during the RF breakdown process, as shown in Fig. 4, which is very different from that of the RF DBDs of atmospheric argon reported in Ref. [7], but is similar to that of the atmospheric helium RF DBDs in Ref. [15] and that of argon-ethanol mixture RF APGDs in Ref. [10].

By comparing the electrical characteristics and the discharge images presented in Figs. 4 and 3, respectively, it can be seen that the breakdown voltage with the Type II plasma generator is lower than that using the Type I plasma generator probably due to the decrease of the dielectric layer thickness in spite of the enlarged gap spacing between electrodes in the Type II plasma generator, since the dielectric constant of glass is about 4 times or more higher than that of argon [16]; for the discharges using the Type II plasma generator, a discharge mode transition occurs with a large discharge voltage drop from...
578.7 V to 510.7 V, which means that the operation window of the discharge current for sustaining the glow mode RF DBDs is from 97.97 to 198.3 mA.

Fig. 4 $V$-$I$ curves of the RF DBDs using different types of plasma generators.

4. Conclusions

In this paper, the discharge characteristics of the atmospheric argon RF DDB plasmas generated with the aid of the atmospheric-pressure HVAC DBDs are studied. The experimental results show that the RF breakdown voltage of atmospheric argon can be reduced significantly with such dual-frequency plasma generator configuration. The large volume, stable and uniform RF DBDs in atmospheric argon is obtained and sustained with turning off the HVAC DBDs after the RF breakdown occurs by using the Type II plasma generator. Further studies concerning the influences of the material and thickness of the dielectric layers, gap spacing between electrodes, size and relative position of the RF and HVAC powered electrodes, etc, on the features of the dual-frequency, atmospheric-pressure, non-equilibrium discharges are necessary.

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References


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<th>I (rms mA)</th>
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<td>(f)</td>
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Table 1. Measured parameters and operation conditions in Fig. 3