

Research progress in the study of atmospheric pressure glow barrier discharge*

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Abstract Atmospheric pressure glow barrier discharge (APGBD) can operate at high pressure, and so vacuum device is not necessary. Furthermore, the produced plasma by APGBD has moderate electron temperature and density besides good uniformity. Therefore, APGBD has extensive potential applications in industry and has been becoming a hot issue in the research of low temperature plasma. In this paper, the main problems in the study of atmospheric pressure glow discharge generated by dielectric barrier discharge, including the experimental setup, judging criterion, discharging conditions, physical mechanisms, and parameter diagnoses, are discussed, and further research prospects of APGBD are proposed.

Keywords: atmospheric pressure glow discharge, dielectric barrier discharge, low temperature plasma.

Low temperature plasma generated by gas discharge has many important application fields, such as growth of thin film^[1,2], modification of material^[3], and plasma display panel^[4]. Moreover, it can be used for ozone generation, removal or degeneration of harmful gas^[5], showing good environmental benefits. However, it is difficult to maintain low pressure in the discharge for plasma generation in industry. Obviously, low temperature plasma generated at one atmospheric pressure is more desirable for industrial application.

Low temperature plasma at high pressure can be generated by corona discharge, dielectric barrier discharge (DBD), and arc discharge. Among these discharges, corona discharge is very weak and the efficiency of radical generation is too low to be suitable for industrial application; contrary to corona, the energy density in arc is too high, and arc discharge may cause damage to work-piece because electron and ion have such a high energy; DBD can easily produce plasma with proper electron temperature and density, so it has extensive application fields in material processing, plasma lighting and environmental protection, etc.

DBD at atmospheric pressure often operates in filamentary mode, and the plasma is not uniform, resulting in the non-uniform disposal of materials and probably causing damage to the surface of the materi-

als. Therefore, the most desirable plasma is produced by large-area uniform DBD, which is called atmospheric pressure glow discharge (APGD) by Okazaki^[2]. Research results show that the efficiency to produce radicals in APGD is much higher than that in filamentary discharge^[6].

In fact, as early as 1933, the research result about APGD has been reported by Von Engel in Germany^[7]. After a stable APGD in inertial gas was reported by Okazaki in 1988^[2], extensive researches have been carried out theoretically and experimentally throughout the world.

1 Typical experimental setup

The typical experimental setup for APGD is shown in Fig. 1. The discharge electrodes are composed of two parallel plates, at least one of which is covered by dielectric (ceramics or glass). The driving frequency ranges from several kHz to a few hundred kHz.

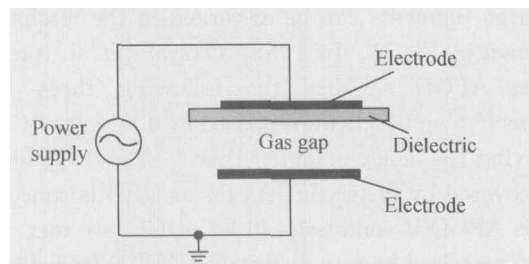


Fig. 1. Experimental setup of parallel planar plates.

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The research results of Okazaki show that APGD is more stable when one of the electrodes has a brush shape^[2,8]. Mangolini used circular planar electrodes to study the radial development of APGD^[9]. Both of the two setups are similar to that shown in Fig. 1 and the applied electric fields are nearly even.

Radu studied APGD in uneven electric field^[10,11]. In his experiment, needle-planar electrodes and rod-planar electrodes were used. Typical setup of uneven electric field is shown in Fig. 2. The upper electrode is like a comb and discharge occurs on the surface of comb electrode^[12,13]. It is called a surface discharge. The working voltage and frequency of surface discharge are similar to those shown in Fig. 1.

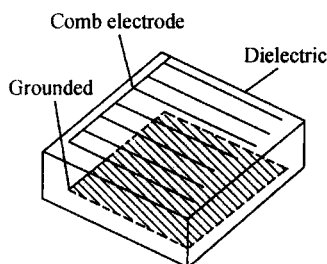


Fig. 2. Experimental setup of surface discharge.

Because of the dielectric, APGD can only operate under ac voltage. A semi-conductor with high resistance has been used by Purwins to substitute for the dielectric^[14-16], so the discharge can operate under dc voltage. APGD is not realized with this experimental setup. Laroussi et al. used water as electrodes to study APGD^[17,18], and they called it resistance barrier discharge because no dielectric was used. In fact, this is DBD of a small dielectric constant.

2 Characteristics and judging criterion of APGD

Generally speaking, it is not APGD if micro-discharge filaments can be discerned in the discharge as shown in Fig. 3. In 1988, Okazaki et al. proposed that APGD satisfies the following three conditions^[19,20]: (1) helium is used as dilute gas; (2) the driving frequency is higher than 1 kHz; (3) electrode is covered by dielectric. As far as DBD is concerned, it is APGD if conditions (1) and (2) are met. They also proposed how to distinguish APGD from filamentary discharge by the waveforms of voltage and discharge current. Fig. 4(a) and (b) give the discharge current waveforms of filamentary discharge and APGD, respectively. In filamentary discharge, dis-

charge pulses are stochastically distributed, and the averaged width of pulse is about tens of nanoseconds. However, in APGD there is only one discharge pulse during each half cycle of the applied voltage and the pulse width is more than $1 \mu\text{s}$ ^[20,21].

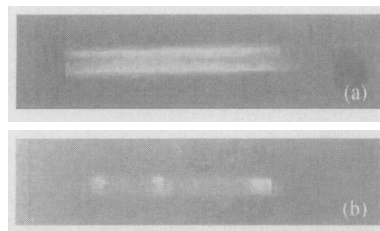


Fig. 3. Photos of APGD (a) and filamentary discharge (b). The exposure time is 0.05 s.

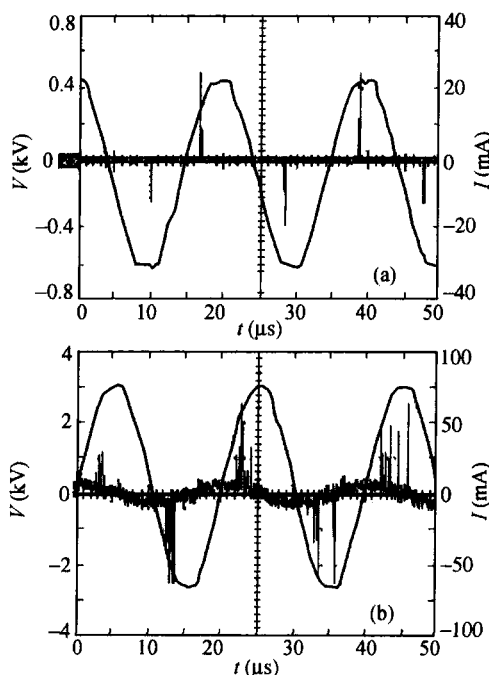


Fig. 4. Waveforms of voltage and current in APGD (a) and filamentary discharge (b)^[21].

If there is only one pulse per half cycle, there will be only two voltage lines in the Lissajous figure because the charge line is only about one microsecond and cannot be discerned in the figure. Therefore, Lissajous figure of filamentary discharge is approximately a parallelogram, but there are only two parallel lines in APGD, as shown in Fig. 5.

Trunec et al. studied the discharge current in atmospheric pressure helium and they found that there are several discharge pulses appearing periodically in each half cycle of the applied voltage and the pulse width is more than $1 \mu\text{s}$, as shown in Fig. 6(a). They called it glow-like discharge^[22]. Fig. 6(b) is

the corresponding Lissajous figure. There are three charge jumps in Lissajous figure, which correspond to the three discharge pulses in Fig.6(a).

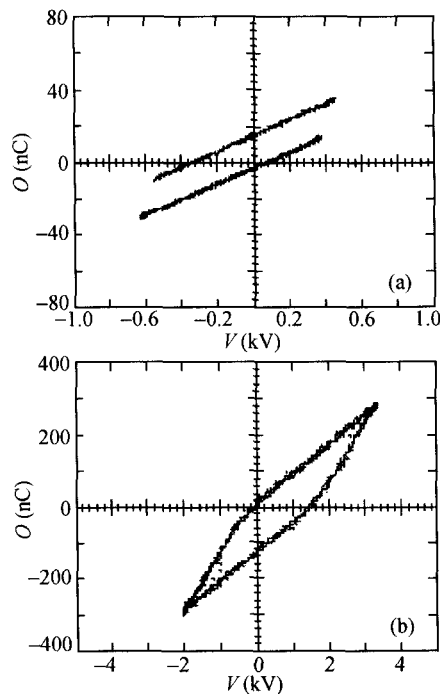


Fig. 5. Lissajous figures of APGD (a) and filamentary discharge (b)^[21].

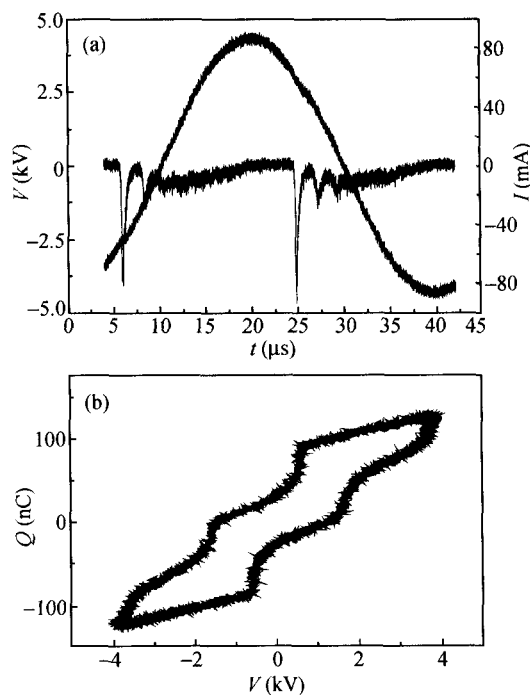


Fig. 6. Waveforms of voltage and current in glow-like discharge (a), and the corresponding Lissajous figure (b).

The characteristic of discharge is a judging crite-

ron of APGD. Consequently, it is very important to obtain the discharge current from the total current measured by a small resistor in series with the discharge electrodes. The current measured by the small resistor includes the displacement current and the discharge current. In order to obtain the discharge current from the total current, the same discharge setups can be connected in parallel. One is ignited, while the other is placed to measure the displacement current. Then, the discharge current can be obtained through deducting the displacement current from the total current. The relations of discharge current $I_d(t)$, memory voltage $V_m(t_0)$ and $V_m(t)$, gas voltage $V_g(t)$ and the applied voltage $V_a(t)$ are^[23]:

$$V_g(t) = V_a(t) - V_m(t), \quad (1)$$

$$V_m(t) = 1/C_{ds} \int_{t_0}^t I_d(t) dt + V_m(t_0). \quad (2)$$

Here, C_{ds} is the equivalent capacitance of the discharge setup. One can obtain $V_m(t)$ and $V_g(t)$ from $V_a(t)$ and $I_d(t)$ by Eqs. (1) and (2), as shown in Fig.7.

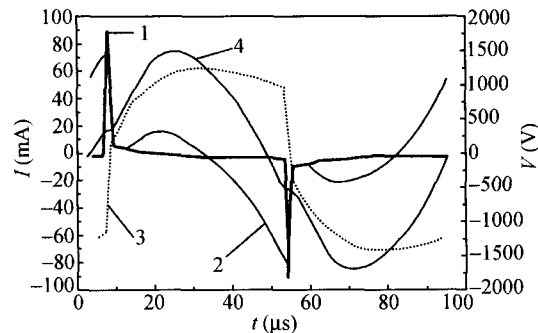


Fig. 7. The relation of $V_g(t)$, $V_m(t)$ and $V_a(t)$ in APGD. 1, $I_d(t)$; 2, $V_g(t)$; 3, $V_m(t)$; 4, $V_a(t)$.

In order to know the discharge uniformity, spatial resolved measurement of the discharge is needed. However, it is difficult to realize spatial resolved measurement of the discharge current, and the spatial distribution of emission can be measured to obtain that of the discharge^[24]. Usually, the discharge is imaged by lens and a diaphragm is placed on the image plane to select the light signal emitted from different places.

Besides the measurements of voltage, discharge current and the distribution of light emission, intensified charge coupled device (ICCD) and streak camera are also used to obtain the discharge photograph with exposure time of nanoseconds^[25]. These photographs are greatly helpful for studying the discharge micro-

scopically and making the discharge mechanism clear. The research results show that there are three regions (cathode fall region, positive column and anode region) from the instantaneous cathode to the anode in APGD, and this is similar to the glow discharge at low pressure.

3 Physical mechanisms of APGD

It is well known that Townsend breakdown mechanism can be used to explain the glow discharge at low pressure. According to this mechanism, instability will occur in the discharge at atmospheric pressure and the discharge will transit to arc or discontinuous spark. Then what is the mechanism of stable APGD?

APGD has attracted the interest of many scientists all over the world, and different theories have been proposed. The research group of Roth in America tried to explain APGD by the ion trapping mechanism^[26]. When the driving frequency is high enough, ions cannot reach the electrode at the interval of half cycle, so they are trapped between the two electrodes. The driving frequency is not high enough to trap the electrons. Under this circumstances, there are many positive charges in the gas gap before the discharge is ignited in each half cycle, and the electric field for breakdown can be lowered greatly, which helps the realization of APGD. According to this theory, if the driving frequency is too low, neither the ions nor the electrons can be trapped, and the discharge can only be ignited under high voltage (over-voltage condition), so the discharge is filamentary. From the ion trapping mechanism, they deduced the relation that the driving frequency ν_0 must be met to realize APGD:

$$\frac{eV_{\text{rms}}}{\pi m_i \nu_{ci} d^2} \leq \nu_0 \leq \frac{eV_{\text{rms}}}{\pi m_e \nu_{ce} d^2}. \quad (3)$$

Here, V_{rms} is the root mean square voltage; m_e and m_i are the masses of electron and ion, respectively; ν_{ce} and ν_{ci} are the collision frequencies of electron and ion, respectively; d is the gas gap width.

The ion trapping mechanism has been doubted. According to this theory, APGD can be realized in arbitrary gas gap width only if the driving frequency is proper, but the experimental results of Wang do not show APGD if the gas gap width is more than 5 mm whatever driving frequency is used^[27]. Based on the simulations, they proposed that the electron density

would reach the critical value before the electron avalanche travels 5 mm, and the discharge will transit to streamer. Consequently, the discharge must be non-uniform.

Research results show that pd value (p is the gas pressure and d is the gas gap width) is a dominant parameter in the breakdown process^[28]. When the pd value is very high, electron avalanche will reach the critical value after a certain distance from the cathode and discharge will transit from Townsend breakdown to streamer. The discharge is not uniform in streamer and APGD cannot be realized. Consequently, only at low pd value can APGD be realized, which means that the gas gap width is less than 5 mm under atmospheric pressure.

At low pd value, the uniform discharge may be Townsend discharge or glow discharge. The main difference between Townsend discharge and glow discharge is that the electric field is almost uniform in Townsend discharge, but in glow discharge, the electric field is distributed mainly in the cathode fall region. The electron avalanche is mainly maintained in the cathode fall region because the electric field is very strong only in this region. Whether it is Townsend discharge or glow discharge at low pd value depends on the working gas. Research results of Massines show that it is glow discharge in helium, while Townsend discharge in nitrogen^[29].

The breakdown mechanisms for both Townsend discharge and glow discharge at low pd value are the same. Electrons are first generated from the cathode surface by bombardment of energetic ions, and these electrons absorb energy from the electric field when they travel from cathode to anode, and then they ionize the neutral particles to create new electrons to form avalanche. Many of these electron avalanches overlap and uniform discharge can be observed. If this kind of discharge can be maintained at atmospheric pressure, stable APGD is realized^[30,31]. Unfortunately, under most circumstances, uniform discharge will transit to arc due to instability. It is commonly considered that heat instability is predominant for the transition from glow discharge to filamentary discharge^[32].

In normal glow discharge, the current density is proportional to the square of pressure, so the discharge current density and quantity of heat are very small under low pressure condition. Consequently,

heat instability is not so noticeable, that is why uniform glow discharge can be easily realized at low pressure. However, at atmospheric pressure, both the current density and quantity of heat are large, so heat instability is remarkable. Suppose that there is fluctuation of current density (slightly bigger) somewhere in the gap, there will be more quantity of heat released by the discharge and the gas temperature is higher than that in other places. The density of neutral particles turns less and the mean free path of electron becomes longer, resulting from heat expansion. The energy of electron increases and more neutral particles can be ionized to produce more electrons at that place. Consequently, the current density there becomes bigger and bigger. The discharge will transit from glow to arc with the increasing current. Accordingly, the uniform discharge will disappear and filaments can be observed.

In order to prevent the transition from glow to arc at atmospheric pressure due to heat instability, many methods have been used. First, proper gases, such as helium, neon and nitrogen, are chosen as working gas^[9]. In these gases, electrons can be generated after each discharge pulse, and these seed electrons can lower the ignition voltage in the next half cycle. Consequently, not too much heat is released and heat instability can be avoided. There are two points of view about the origin of seed electrons. One believes that it comes from collisions between metastable particles or Penning ionization, and the other believes that it is produced by the collisions between metastable particles and dielectric surface. Therefore, a certain concentration of seed electrons is needed to obtain APGD, and the driving frequency needs to be high enough to obtain stable APGD. This explains why the driving frequency of more than 1 kHz is used for APGD in helium, pointed out by Okazaki^[2].

Using needle-plane electrode configuration, Takaki et al. found in atmospheric pressure nitrogen discharge that the discharge starts from glow discharge and the discharge current increases with time, at last the discharge transits to arc when an overvoltage is applied on the two electrodes^[33]. Therefore, the discharge current should be restricted to prevent the transition from glow to arc. In 1993, by restricting the increment of discharge current, APGD was realized in helium, nitrogen, argon and even in air^[20]. In the experiments, a fine mesh wire was placed between the electrode and the dielectric as a re-

sistor to restrict the increment of discharge current. Through restricting the discharge current, APGD could be realized even without seed electrons, so no driving frequency was limited. They even obtained APGD at line frequency. Unfortunately, Trunec et al. pointed out that APGD at line frequency is not very stable in argon and nitrogen except in helium^[34]. Yang^[35] used copper oxide as a resistor to restrict the increment of discharge current, and APGD was obtained in the configuration of needle-needle electrodes in air and methane. Similarly, Purwins et al. used semiconductor and found that it was glow discharge under low pressure^[14,36]. There were not any dielectrics in their experiments and the discharge could be maintained under dc voltage. However, they were only interested in the dynamics of pattern formation. Laroussi et al. used water as high impedance electrode to study APGD^[17,18].

In the experiments of APGD, flowing gas is often used^[37]. The flowing of gas helps to take away heat and the transition from glow to arc resulting from heat instability can be avoided. Consequently, uniform glow can be easily observed in flowing gas.

Experimental results also show that humidity of working gas and trace impurity can affect APGD greatly. Stable APGD in helium can transit to filamentary discharge if trace water vapor or oxygen or carbon dioxide is filled into the working gas^[38].

4 Diagnosis of plasma parameters

Much progress has been made in the study of APGD. However, one needs to know the plasma parameters to facilitate its application in industry. It is easy to diagnose these parameters at low pressure, but there are still many difficulties for atmospheric pressure plasma. Diagnosis of plasma parameters of APGD is still at the starting stage.

Langmuir probe has been widely used in diagnosis of plasma parameters, but under atmospheric pressure, the condition that the mean free path of electrons is longer than the thickness of plasma sheath cannot be met, therefore, Langmuir probe cannot be used at atmospheric pressure.

Microwave interference method is valid for Coulomb collision dominated plasma. However, the ionization degree in APGD plasma is very low, and so the predominant collision is between electron and neutral particle. Therefore, microwave interference

method cannot be used to diagnose plasma parameters of APGD.

Laser Thomson Scattering (LTS) is a precise method for diagnosis of plasma parameters^[39]. The fundamental theory of LTS is: charged particles in the plasma will oscillate with the electric field of electromagnetic wave when laser enters the plasma. The oscillating charged particles can give out secondary electromagnetic wave, which is scattering wave. The scattering wave comes mainly from oscillating electrons because the mass of ions is much larger than that of electrons. Information of electron temperature and plasma density is contained in the scattering wave. Through analyzing the scattering wave, one can obtain the plasma parameters precisely. This method is applicable for the diagnosis of high temperature plasma. However, the scattering wave from neutral particles cannot be neglected at atmospheric pressure, and LTS is not suitable for plasma parameters diagnosis of APGD. Even though the scattering problem from neutral particles can be resolved in the future, the prospect of industrial application of LTS is not too prosperous because of the expensive equipments.

Plasma parameter diagnosis by emission spectroscopy is a feasible method for APGD. The broadening of argon spectral line comes mainly from the interacting between emission atom and electrons or ions. It is a kind of secondary broadening, so the Stark broadening of argon spectral line can be used to obtain the electron density^[40]. High-resolution spectrometer is needed to measure the Stark broadening width directly. We proposed a method to obtain the Stark broadening by separating different spectral profiles^[41]. The profiles of spectral line resulting from different broadening mechanisms are not the same. The profile of Stark broadening is Lorentzian, and that of instrumental broadening is Gaussian. Through convolution of Lorentzian profile and Gaussian profile the experimental profile is fitted, then de-convolution process is conducted to separate the Lorentzian profile to calculate the width of Stark broadening, and so the electron density of plasma can be obtained. The results show that the electron density in the filaments is in the order of 10^{15} cm^{-3} , and the electron density of APGD has not been studied by far.

5 Perspectives

The prospect of APGD in industrial application is

attractive, but there has not been large-scale application in industry because the mechanism of stable APGD is not very clear, and successful research results of APGD are only related to helium. Further study should emphasize on the mechanism of APGD, especially that in electrode configuration of nonuniform applied electric field such as surface discharge, transition condition from glow to streamer and diagnosis of plasma parameters. If more progress is made in these research aspects, the application of APGD will be extensive and stirring.

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