Spectroscopic Study of a Radio-Frequency Atmospheric Pressure Dielectric Barrier Discharge with Anodic Alumina as the Dielectric∗

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Abstract This paper presents the fabrication and a spectroscopic study of a stable radio-frequency dielectric barrier discharge (RF DBD) in Ar with a novel dielectric, anodic alumina, at atmospheric pressure. Dielectric electrodes are fabricated from commercially available low cost impure aluminum strips by a two-step anodization process in 0.3 M solution of oxalic acid. The discharge is found to be stable with excellent spatial uniformity for the RF input power range of 30∼80 W. Excitation and rotational temperatures measured in the experiment range of 1472∼3255 K and 434∼484 K, respectively, as the input power changes from 30 W to 80 W. These temperature ranges are suitable for surface modification applications.

Keywords: radio frequency atmospheric pressure glow discharge, dielectric barrier discharge, anodic alumina dielectric

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1 Introduction

Recently, atmospheric pressure glow discharge (APGD) sources have attracted considerable interest, primarily because they offer the distinct advantage of chamberless apparatus for various technological applications [1]. These sources include the cold plasma torch, atmospheric pressure plasma jet (APPJ) [2], micro-hollow cathode discharge (MHCD) [3,4], one atmosphere uniform glow discharge plasma (OAUODP) and dielectric barrier discharge (DBD) [5]. The non-equilibrium character of glow discharge plasmas combined with high pressure operation offers immense potential for applications such as low temperature surface modification, bio-sterilization, plasma display panels, pollution control, excimer ultraviolet lamps and deposition of thin films [6∼10].

A stable and spatially homogeneous large area of glow discharge is required for various industrial applications. The stability of APGDs is limited due to their tendency to glow-to-arc transition. Plasma stability is robust in RF DBD [11]. SHI and KONG [12] demonstrated the generation of a stable, large area atmospheric Ar glow discharge over a large range of discharge current using RF DBD. SHI and KONG [13] also studied the basic characteristics of mode transition with bare and dielectrically insulated electrodes in radio frequency Ar discharge. The presence of dielectric barriers, unlike bare electrodes, results in a large discharge for both alpha and gamma modes with gradual mode transition. LI et al. [14] generated an atmospheric argon glow discharge with RF DBD by using water cooled copper electrodes covered with quartz sheets and a 5.5 mm gap. The results show that secondary electron emission might be responsible for the sustainable glow discharge in the large gap RF DBD.

Porous oxide films can be obtained by anodizing aluminum in acidic electrolytes. Wet chemical processing eliminates dielectric adhesion problems when dielectric barrier electrodes are fabricated from metals. The growth kinetics and self organization of ordered pore arrays have been widely discussed [15,16]. CHO et al. [17] presented a DBD reactor with 2 μm thick anodic porous alumina barrier developed by anodizing cylindrical aluminum rods in H2C2O4 at 15°C. The DBD was generated at 20 kHz, 460∼510 V range in 50 torr Ne with 5 mm gap spacing. The device was proposed for the generation of ultraviolet radiation, lighting and displays. PARK et al. [18] fabricated microdischarge devices with 10 μm thick nanoporous alumina dielectric produced by wet chemical processing operated at 20 kHz and 650 V in 700 torr Ne and 800∼850 V in air at atmospheric pressure. The reported system yields devices that are exceptionally robust, long-lived, lightweight, inexpensive and suitable for processing over large areas. KAWASAKI [19] generated a stable RF DBD at 4.6 kV with anodic porous alumina dielectric in air at atmospheric pressure. Pure aluminum (99.99%) was anodized at a constant DC voltage of 24 V in 550 mL H2SO4 solution. The optimization of anodizing conditions from a viewpoint of stable DBD generation was also presented.

In this paper, the fabrication of dielectric electrodes from anodization of commercially available low cost impure aluminum for the development of a stable RF DBD is reported. The primary objective of this work is to generate and study the optical characteristics of RF Ar atmospheric glow discharge sustained between two parallel anodized aluminum electrodes.

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2 RF DBD reactor

2.1 Anodization of aluminum electrodes

Impure aluminum strips (30 mm × 30 mm) were first etched in 5% NaOH at 60 °C for 30 s. The two step anodization was carried out to obtain a porous anodic alumina film on the surface of the aluminum. The schematic diagram of anodization setup is shown in Fig. 1. During anodization, an aluminum piece was connected to the negative terminal while the aluminum electrode to the negative terminal of a DC power supply (Tenma 72-7245). The first step anodization was carried out in a 0.3 M oxalic acid solution. The anodization time was 30 min with a constant voltage of 30 V. The solution was placed in an ice bath to maintain a temperature below 20 °C. After the first step, the oxide layer of the sample was removed by etching for 15 min etching in 1.5 wt% H$_2$CrO$_4$ + 6 wt% H$_3$PO$_4$ solution at 60 °C. After removing the oxide layer, the second step anodization was done in 0.3 M oxalic acid for 2 h. This step developed an alumina film on the aluminum strip connected to the positive terminal of the power supply. The anodized aluminum strips were used as the electrodes of RF DBD.

2.2 Plasma source and the diagnostic setup

A schematic illustration of the experimental setup and electrode assembly is shown in Fig. 2(a) and (b), respectively. The discharge was created in an open air electrode assembly without isolating from the laboratory air. The gap between electrodes, mounted on Teflon, was kept at 1.2 mm using glass spacers. The discharge was powered by a 13.56 MHz RF generator (CX600AS/13.56 MHz) through a matching network (Match Pro CPM-1000). Plasma was generated at atmospheric pressure by feeding industrial grade Ar gas with a flow rate of 7 standard liters per minute (SLM). The ability of He to produce a stable glow discharge at atmospheric pressure is well known due to its high thermal conductivity and low breakdown voltage [20]. However, it is economically desirable to use a cheaper gas like Ar, as compared to He in APGD applications. The visible emission spectrum from the discharge was obtained by an Avantes spectrometer (Avaspec-3648) having a charge-coupled device (CCD) detector array with 1200 grooves/mm gratings. The discharge pictures were taken with a digital camera (Kodak C 913).

3 Results and discussions

3.1 Anodization of aluminum electrodes

Fig. 3 shows a scanning electron microscopy (SEM) image of the top surface of the porous anodic alumina, anodized for 2 h in a 0.3 M solution of oxalic acid at a constant voltage of 30 V. The pores appear to have no planar ordering, and they are inconsistent and of noncircular shapes. The self organization is disrupted by enough pore growth to merge with adjacent pores, destroying the hexagonal array. It is concluded that the experimental conditions were unfavourable for the formation of ordered pore arrays, i.e., a low anodization voltage and impure aluminum. The mechanical stress due to the volume expansion of aluminum during the oxidation process produces a repulsive force between the pores during growth, leading to self-organized hexagonal pore arrays [15]. Low anodization voltage is suggested to be responsible for the formation of non-uniform pore structures because the volume expansion during aluminum oxidation occurs only at high anodization voltage [21]. Furthermore, the surface roughness of the aluminum samples also affects the formation and distribution of pores. The high roughness of the surface results in the quick formation of a barrier oxide as well as pores at depressions. The pores that appear at such depressions in an early stage will grow in front of the other pores [16]. A SEM micrograph (Fig. 3) also reveals irregular holes of micrometer size, appearing not to be through-thickness at some locations, which are in good agreement with observations.
for impure aluminum reported elsewhere \cite{22}. The reported growth rates of porous anodic alumina for impure aluminum, anodized in oxalic acid ranging from 40 nm/min to 60 nm/min \cite{22}, indicate that our porous anodic alumina films have a thickness between 5 \( \mu \text{m} \) and 7 \( \mu \text{m} \).

3.2 Glow discharge and excitation temperature

A white purplish glow discharge spread throughout the volume at 30 W RF input power. Visual observations show that the discharge is in stable \( \alpha \)-mode with excellent spatial uniformity. If the input power is increased from 30 W onwards, the Ar RF DBD remains volumetric and the optical emission increases with increasing RF input power up to 80 W. If the input power is increased beyond 80 W, the RF DBD in the \( \alpha \)-mode transforms to that in the \( \gamma \)-mode. In the input power range of 30\(\sim\)80 W, the discharge is found to be operating in a stable \( \alpha \)-mode without any sign of arc or filaments. The photograph of DBD in the \( \alpha \)-mode recorded at 50 W is presented in Fig. 4.

Compared to RF APGD jets \cite{2}, the argon lines in RF DBD are more in number, thus indicating active underpinning plasma chemistry. For example, optical emissions at 811 nm, 826 nm, 841 nm, and 843 nm were found to be weak in an Ar RF APGD jet \cite{2}. In an Ar kHz plasma needle \cite{23}, the line observed at 750 nm was the strongest among all lines, including the lines at 697 nm and 772 nm, thus exhibiting an interesting contrast to the spectrum of Fig. 5.

In this study, Ar I emission lines at 706.78 nm, 714.74 nm, 727.37 nm, 750.48 nm and 826.37 nm are selected to determine the excitation temperature using Boltzmann’s plot method \cite{24}. Fig. 6 presents a Boltzmann plot of the selected Ar I spectral lines. The variation of excitation temperature with the input power at a fixed gas flow rate of 7 SLM is presented in Fig. 6. As the input power of the RF DBD increases in the range of 30\(\sim\)80 W, the excitation temperature rises from 1472 K to 3255 K. As the RF input power rises, the electrons gain more energy from the applied electric field. Therefore, the excitation temperature increases due to more energetic electrons. The excitation temperature of the Ar RF DBD is similar to that in other Ar RF atmospheric plasma \cite{25} but lower than that in low pressure Ar glow discharges \cite{26}. At atmospheric pressure, the electron-neutral collision frequency is very large compared with the small order of mean free paths. As a result, the electrons are able to accumulate a very small amount of energy, which leads to low excitation temperatures as compared to low pressure Ar glow discharges.

Due to the frequent collisions among heavy particles at atmospheric pressure, it is known that the rotational temperature is nearly equal to the gas temperature. The optical emission spectrum of OH was used to...
estimate the gas temperature. By comparing the shape of the measured OH line around 309 nm with LIFBASS simulation data [27], the gas temperature was obtained.

The gas temperature of Ar RF DBD, as a function of the input power, varies from 434 K to 484 K, as shown in Fig. 7. The gas temperature increases with increasing input power, which is attributed to more energy transfer from heated electrons to heavy neutral atoms. The range of measured gas temperature suggests that the plasma can be readily applied to material surface modifications [28].

4 Conclusions

In conclusion, a stable Ar RF DBD is developed by anodizing, commercially available low cost impure aluminum strips, in a 0.3 M solution of oxalic acid. The discharge is found to be stable with excellent spatial uniformity in the RF input power range of 30 W ∼ 80 W. The excitation and rotational temperature of the RF DBD increases respectively from 1472 K to 3255 K and from 434 K to 484 K for the aforesaid power range. This gas temperature range is suitable for surface modification.

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References


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