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A high-activity nitrogen plasma flow source for deposition of silicon nitride films

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ABSTRACT

We report a tubular plasma source that is capable of creating high-activity nitrogen plasma flow at low pressure. The high-activity nitrogen plasma was produced by a continue low-frequency discharge, in which an intensive pulsed discharge was observed when the electrode was polarized by the positive voltage. Excited at 10 to 115 W, the plasma source allows loading the power density as high as ~80 W/cm^3 to the plasma, producing high-activity nitrogen plasma with a maximum dissociation degree of nitrogen larger than 10%. Based on the tubular plasma source, a special system of plasma enhanced chemical vapor deposition has been developed for deposition of low H-content amorphous hydrogenated silicon nitride (*a*-SiN_x:H) films at room temperature.

1. Introduction

Recently, electric discharges with highly constricted configuration have received much attention because of the potential for a series of novel applications. For example, microdischarges spatially confining the plasma to dimensions of 1 mm or less enable generation of stable glow discharges in a wide variety of gases at atmospheric pressure with as high as 10^{15} cm^{-3} electron density, which is considered applications as plasma cathodes [1]. Atmospheric pressure plasma jet (APPJ) is a facile tool for production of plasma beams and has received much attention due to its prospect in low-temperature processing of materials [2–14]. However, it is generally difficult to deposit high quality of films using an APPJ. It is common knowledge that discharge conditions, such as the working pressure, the discharge configuration, and the excitation way, determine the properties of plasma. At low pressure, the mean free path of molecules and Debye length of electrons are extended to a scale closed to the diameter of APPJ's dielectric tube. Therefore, a low-pressure tube source, which is analogue of APPJ, is particularly interesting to produce high activity of nitrogen plasma for synthesis of nitride films, such as GaN, AlN, and SiN_x, which are important in optoelectronic and photovoltaic applications. For example, Czerwiec et al. designed a cylindrical source of inductively coupled plasma (ICP) for generation of plasma beams at low pressure [15,16]. The plasma source is capable of creating high density of nitrogen plasma with an association degree higher than 10%. Henriques et al. studied the nitrogen dissociation of a surface wave driving N_2–Ar discharge in a 0.75 cm quartz tube [17]. Kessels et al. [18–20] developed an expanding thermal plasma source for high-rate deposition of amorphous hydrogenated silicon-nitride (*a*-SiN_x:H) films with plasma enhanced chemical vapor deposition (PECVD) method. In addition, some PECVD systems were developed on the basis of hollow cathode discharge [21].

In this study, we report a low-frequency (20–80 kHz) driving tubular plasma source operating at the pressures ranging from 1 to 200 Pa. This plasma source can be sustained at an intensive discharge state consisting of continue discharge and periodic pulsed discharge when using nitrogen as the working gas, thus creates a high activity of nitrogen plasma. This tubular plasma discharge is rather different from a hollow cathode plasma discharge because we used a dielectric tube confined the plasma and an addition metal rod as the transient cathode. Using the high activity of nitrogen plasma, low H-content amorphous silicon nitride (*a*-SiN_x:H) films were deposited at room temperature in a relatively high rate.

2. Experimental methods

The nitrogen discharge was produced on an experimental setup schematically shown in Fig. 1(a). Fig. 1(b) shows the typical photographs of the bright discharge operating at the given pressures. The plasma source consisted of a quartz tube with inner diameter of 4.0 mm and a 2-mm thick electrode. The electrode was made of stainless steel and driven by a low-frequency AC generator (20–80 kHz) using the vacuum chamber as the ground. The power rating of AC

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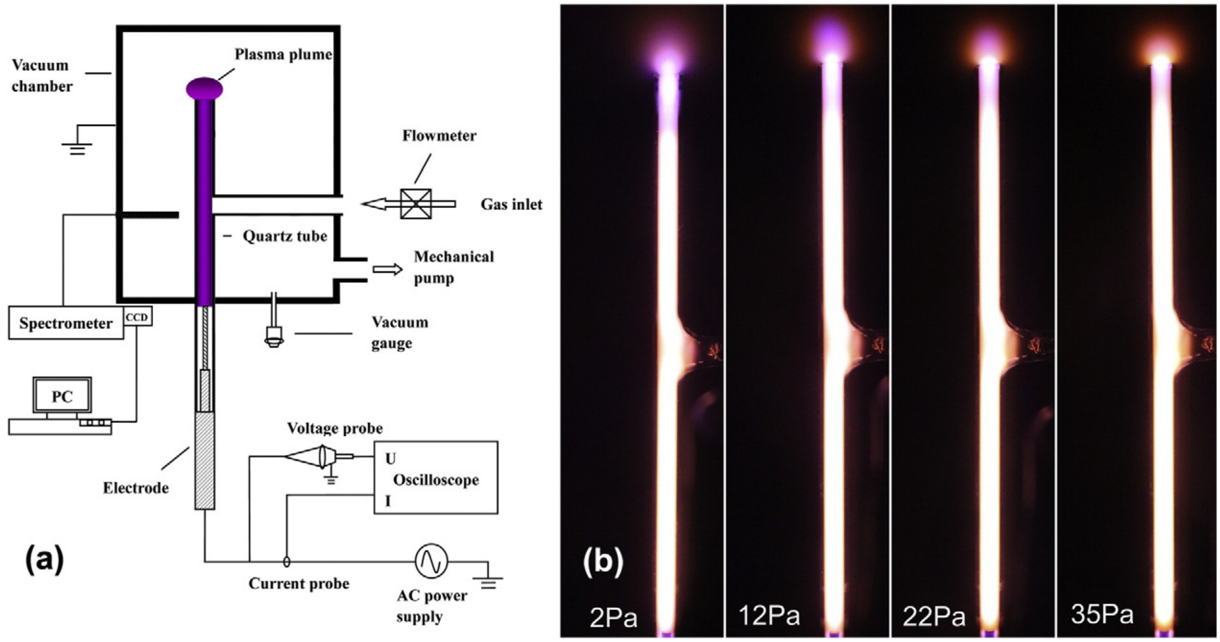

Fig. 1. (a) Schematic experimental setup. (b) Typical photographs of the discharge operating at the given pressures.

generator was 500 W. Before discharging, the chamber was pumped to be better than ~1.0×10^{-3} Pa, and then injected with 12 SCCM 99.999% nitrogen through a gas inlet on the sidewall of the tube. To deposit *a*-SiN_x:H films, a PECVD system was specially designed using a quartz co-axially double tube to replace the single tube for generation of nitrogen plasma. The inner diameters of double quartz tube are 4 and 8 mm, with ~1 mm thick walls, thus pure nitrogen and Ar-diluted 5% SiH_4 are separately fed through the inner and the outer tubes.

Optical emission spectroscopy (OES) was used to study the plasma in the tube. The optical emission was spectrally analyzed in the wavelengths ranging from 200 to 1100 nm using a spectrometer (Acton Research Spectra, Pro-2500i) with a 1200 mm^{-1} grating. The slit entrance was set to ~10 μm, providing a spectral resolution of ~0.05 nm. The electrical characteristics of discharge were measured with a 1:1000 high-voltage probe (Tektronix P6015A 1000×, 3.0 pF, 100 MΩ) and a current probe (Pearson 6600 0.1 V/A) via a digital storage oscilloscope (Tektronix TDS 2012B, 100 MHz, 1 GS/s). In addition, a Langmuir double probe placed at the center of plume, which was ~5 mm away from the nozzle, was used to measure electron density and temperature in the plume.

3. Results and discussion

3.1. Discharge behavior of the tubular plasma

An application of high voltage to the electrode led to breakdown of the gas and creation of plasma confined in the tube. With an increase in input power, the plasma column in the tube gradually expanded along the tube. At a critical voltage, the discharge abruptly produces dazzling-white plasma filling the entire volume, and then a 10–20 mm long plasma plume appears at the tube nozzle, as shown in Fig. 1(b). Fig. 2 shows the typical discharge waveforms of the dazzling-white plasma excited at 115 W. With the sinusoidal excitation, the pattern of the discharge voltage was heavily deformed, and the discharge current exhibited a deformed sinusoidal wave containing the pulsed discharge with extremely high peak current, thus producing a pulse power more than 1 kW within a typical pulse-duty less than 1 μs. The discharge events were highly periodic and the pulsed discharge occurred always at a voltage-rising phase, though the variation in peak current appeared from one cycle of the applied voltage to another. Similar current pulses have been observed in an APPJ excited by AC power supply [4]. In fact, the diffuse plume outside from the tube is very similar to the plume seen when helium APPJ is emerging into helium atmosphere [22]. Differing from the discharge of the APPJ, there is no pulse was observed when the electrode was under the negative polarization. On the other hand, the phase angle between the discharge current and the applied voltage was much smaller than 90° (35°–60°), suggesting the resistive discharge is dominant. The tubular source was very stable and can be sustained for many hours. The plasma source has the advantage in loading sufficient power density to excite

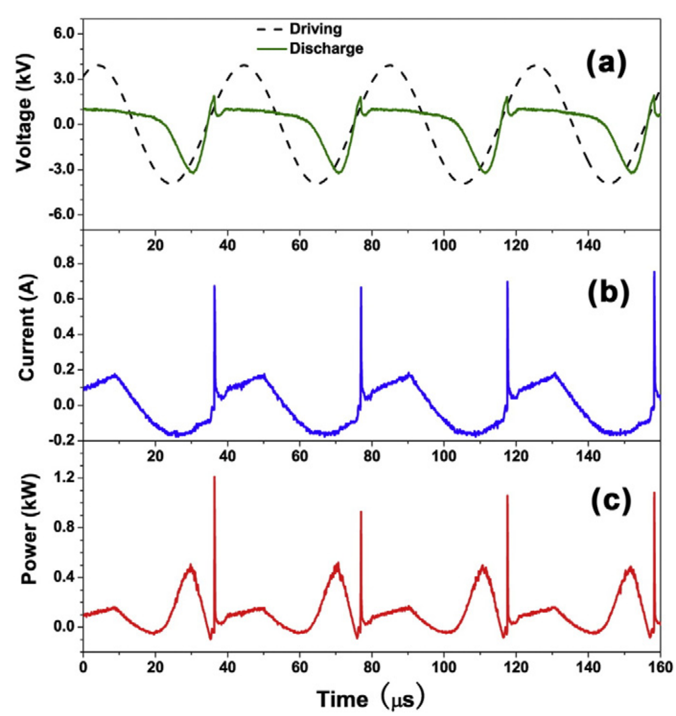

Fig. 2. Typical discharge waveforms, (a) voltage waveforms before and after discharge, (b) current waveform, (c) instantaneous power. The discharge was operated in a 315 mm long quartz tube driven at 12 Pa, 25 kHz, and 115 W.