論 文 内 容 の 要 旨

Plasma technologies attract great attention owing to fast growing interests in variety of plasma-based applications in research and industry with consideration for the sustainable development of humankind. In this work, systematic research and development in designing novel pulsed power generators for energy efficient pulsed plasma discharge methods at both low-pressure and atmospheric-pressure have been carried out. The main purpose of this study is to design novel pulsed power generator circuits with high energy conversion efficiency and wide range of controllable parameters with examining how they perform in actual application to pulsed DC plasma chemical vapour deposition (CVD) of carbon thin films and to atmospheric-pressure plasma jet (APPJ).

In the first part of research, low-pressure pulsed plasma discharge for the synthesis of carbon thin films was investigated. A pulsed DC plasma chemical vapour deposition (CVD) system with a compact vacuum chamber using a simple configured pulsed DC power supply was developed. The first pulsed DC power supply was a conventional half-bridge circuit consisting of a couple of metal-oxide-semiconductor field-effect transistors (MOSFETs) for switching a high voltage direct current (HVDC) like as a push-pull amplifier. The pulsed DC discharge was examined with variation of operation parameters as well as source gas materials. Diamond like carbon (DLC) films, as an example of carbon thin films, were deposited on silicon (Si) substrates from acetylene (C_2H_2) or carbon-monoxide (CO) at two distinct temperatures. The fabricated films were characterized by Raman spectroscopy, FTIR spectroscopy, and scanning electron microscopy (SEM). The results reveal that the films prepared from CO gas exhibited soft conductive DLCs with a large number of graphite like structures whereas mostly insulating hydrogenated amorphous carbon DLCs were obtained from C_2H_2 gas. The deposition rate from CO was lower than from C_2H_2 due to etching by oxygen produced from the source gas. As the substrate temperature increased, progressive graphitization was incorporated to the films deposited from both gases. By using the developed pulsed DC plasma CVD system, the energy efficiency for CVD as the ratio of growth volume to energy consumption was dramatically improved with some distinctive differences in film properties as compared to our previous RF plasma CVD. While the results showed many advantageous features of pulsed DC plasma CVD for replacing conventional RF plasma CVD, it was confirmed that the push-pull operation for the capacitive load such as an electrode caused loss of energy at every pulsed discharge with resulting in limitation of operation frequency.

A novel pulsed-DC generator for capacitively coupled plasma (CCP) discharge was developed to improve energy conversion efficiency and operation frequency for further advanced pulsed plasma discharge processes. The circuit design, the system assembly, and the operating tests of the high-frequency and HV pulsed DC generator (PDG) for CCP discharge have been investigated. For such the capacitive loads, it is challenging to obtain sharp rectangular pulses with fast rising and falling edges, requiring intense current for quick charging and discharging. In addition, the loss of energy usually occurs from the capacitive loads at every termination of pulse. The requirement of intense current and loss of energy generally limit the pulse operation frequency. To overcome these constraints, a new type of PDG consisting of a pair of half- resonant converters and a constant current-controller circuit connected with high-voltage solid-state power switches was developed that could deliver almost rectangular high voltage pulses with fast rising and falling edges for capacitively coupled plasma discharge. A prototype of the PDG was assembled to modulate from a HVDC input into a pulsed HVDC output, following an input pulse signal and a set current level. The pulse rise time and fall time are less than 500 ns and 800 ns, respectively, and the minimum pulse width is 1 μs. The maximum voltage for a negative pulse is 1000 V, and the maximum repetition frequency is 500 kHz. During the pulse on time, the plasma discharge current is controlled steadily at the set value. The half-resonant converters in the PDG perform recovery of the remaining energy from the capacitive load at every termination of pulse discharge. The PDG performed with high energy efficiency of 85% from the HVDC input to the pulsed DC output at a repetition rate of 1 kHz, and with stable plasma operation in various discharge conditions. The results suggest that the developed PDG can be considered to be more efficient for plasma processing by CCP.

To examine the performance of novel pulsed-DC discharge system consisting of the developed PDG and the custom-made compact vacuum chamber, hydrogenated amorphous carbon (a-C:H) films were deposited on silicon (Si) substrates using a high-repetition microsecond-pulsed DC plasma chemical vapor deposition (CVD) from acetylene (C_2H_2) at a gas pressure of 15 Pa. The plasma discharge characteristics, hydrocarbon species, and the microstructure of the resulting films were examined at various pulse repetition rates from 50 to 400 kHz and a fixed duty cycle of 50%. The optical emission spectra confirmed the increase in electron excitation energy from 1.09 to 1.82 eV and the decrease in the intensity ratio of CH/C_2 from 1.04 to 0.75 with increasing pulse frequency, indicating the enhanced electron impact dissociation of C_2H_2 gas. With increasing pulse frequency, the deposition rate gradually increased, reaching a maximum rate of 60 nm/min at 200 kHz, after which a progressive decrease was noted, whereas the deposition area was almost uniform for all the prepared films. Clear trends of increasing sp^3 content (amorphization) and decreasing hydrogen (H) content in the films were observed as the pulse repetition rate increased, while most of the hydrogen atoms bonded to carbon atoms by sp^3 hybridization rather than by sp^2 hybridization.

For further evaluation of the novel pulsed-DC discharge system, argon (Ar) gas discharge experiments were performed under a variety of discharge conditions, and plasma conditions were evaluated using optical emission spectroscopy (OES). The developed system can perform glow discharge by employing nearly rectangular, unipolar, and high-voltage negative pulses with a wide variation of voltage amplitudes and a set discharge current, pulse repetition rate, and duty cycle. Plasma conditions could also be tuned by varying the pulse parameters. Diamond-like carbon (DLC) films were deposited on silicon (Si) substrates from acetylene (C_2H_2) with a variation of gas pressure, applied voltage, and discharge current. The Raman spectra confirmed the DLC properties of the deposited carbon films. All DLC films demonstrated highly insulating properties, and the amorphization increased with a decrease in gas pressure, voltage, and discharge current. With intention of applying the develop system to growth of graphene, ultrathin pyrolytic carbon (PyC) films have been produced on glass substrates using nano-thickness metal catalysts from C_2H_2. While graphene growth has not been confirmed at the examined conditions, the ultrathin PyC films exhibited conductive and semitransparent properties. These results revealed that the developed pulsed-DC discharge system for plasma CVD is applicable for the synthesis of insulating hard DLC coating as well as soft-conducting ultrathin carbon thin films.

The experimental results of the first part are summarized as follows. The original developed pulsed DC plasma CVD system with a compact vacuum chamber and a simple half-bridge pulsed power supply was advantageous due to energy-efficient deposition. However, the original system had the limitation of the modulation of pulse parameters such as high repetition rate, fast rise and fall time, and wide control of pulse width. A novel pulsed plasma discharge system was successfully developed which was more versatile and favorable for the operation of high frequency with fast rising and falling edges. Plasma discharge characteristics, the deposition of films and its properties can be widely modified by the developed novel system, which may open new possibilities in etching, ion implanting, and sputtering applications for the material processing.

In the second part of research, atmospheric-pressure pulsed plasma discharge for plasma jet was investigated. Atmospheric-pressure plasma jet (APPJ) is a kind of small dielectric barrier discharge (DBD) device that can be run under atmospheric-pressure in the ambient air also with various gases and it is technically simple and low-cost process because no vacuum systems is required. In the APPJ system, the power supply is considered one of the significant components, as it plays a key role on the discharge characteristics of the APPJ. Among various types of power supplies, pulsed generator is considered as it can deliver high instantaneous power with respect to low average power, offer fast rise time and high repetition rate which are advantageous to control the production of high electron density and more active plasma species.

For operation of APPJ, alternative current (AC) HV power sources of sinusoidal wave at lower frequency than a several 10s of kHz are widely used because it is easy to boost the voltage by using a transformer. In most cases, the APPJ is operated at a fixed frequency and a fixed voltage. For research and development of APPJ technology, however, it is important to examine the effects of operation frequency and waveforms on the plasma properties and processed results. For operation of DBD, the voltage increasing rate, decreasing rate, pulse width and pulse interval are crucial parameters. For control of APPJ process induced in gas jet, the operation frequency is also an important parameter. In the case of conventional sinusoidal wave ACHV, with the variation of operation frequency and peak voltage, the voltage increasing rate, decreasing rate, pulse width and pulse intervals automatically varies. Because of the sinusoidal property, it is impossible to control the important discharge parameters intentionally. For advancing research on APPJ, it is necessary to develop a novel pulse power source which enables independent control of frequency, voltage, and waveforms.

A high-voltage high-frequency impulse generator (IG) for atmospheric-pressure plasma jet (APPJ) was developed using dual resonance pulse transformer (DRPT) and solid-state switches. The DRPT performs not only to boost the voltage from the primary circuit to the secondary circuit but to recover the energy from the secondary circuit to the primary circuit. At the appropriate condition, the energy conversion efficiency is dramatically improved and it enables high frequency operation with a same pulse waveform. It has been revealed, in this work, that the conditions for total transfer of energy (neglecting dissipation) from primary capacitor to secondary capacitor are: the resonant frequency of the primary circuit for open secondary circuit should be equal to the resonant frequency of secondary circuit for open primary, and the transformer coupling coefficient should be 0.6. Under these conditions, the two normal modes of frequencies of the coupled primary-secondary circuits have in the ratio of 2:1. In the optimized DRPT, the high voltage is not proportional to the number of turns ratio as in an ordinary transformer but formed by resonance. If the capacitance of the secondary circuit is very small compared to the primary capacitor and the inductance of secondary coil is very large compared to the primary coil, the primary voltage is stepped up to a high value in secondary.

The developed generator is compact and has convenient control for voltage and frequency selection. It can deliver high-voltage impulses (peak to peak) up to 11 kV from a DC voltage of 500 V. The pulse operating frequency range is 0.5-100 kHz. The fixed pulsed width is approximately 4 μs with the pulse rise-time and fall-time are almost 2 μs. The DRPT-IG is tested by producing a stable room-temperature atmospheric pressure plasma plume using helium (He) gas flow in a plasma jet device designed as cylindrical configuration of DBD with single electrode configuration. It is probable that this work may stimulate the further development of pulsed generators for atmospheric-plasma discharge processes.

Atmospheric-pressure plasma jets designed as cylindrical configuration of DBD was operated using the developed DRPT-IG and the plasma was examined by means of electrical and optical characteristics using He as working gas. The power source is connected to two different types of single-electrode configurations such as strip-electrode and wire-electrode for the production of plasma jet with various experimental conditions. Wire-electrode configuration shows a strong impact on plasma jet operation with the variation of frequency at a fixed voltage and gas flow rate rather than strip-electrode. The optical spectral analysis for wire-electrode APPJ confirmed the existence of active species such as NO, N_2, N_2^+, He, O and OH in the plasma plume. The spectral intensities of these species increase with the pulse frequency and become most dominant at 100 kHz. This results reveal that the high frequency operation of plasma jet by the developed DRPT-IG is preferable for generation of more reactive species. However, further investigations are required, which are still ongoing by this approach.