Gas Temperature and Cu Vapor Density in an Impulse Discharge

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The laser scattering techniques are very powerful for the diagnostics of impulse discharges. Characteristics of gas temperature, density of Cu atoms from electrodes and electron density in and near an impulse discharge channel at an air pressure of 66.7hPa were investigated. The gas temperature took much higher values in the constricted regions of the channel, especially in the two regions near the tips of anode and cathode along the positive column. At the transition from glow to arc, the gas temperature and the densities of Cu atoms and electrons in the discharge channel became high with a rise of discharge current, and reached maximum values of 4800K, $9 \times 10^{18} \text{ m}^{-3}$ and $8 \times 10^{23} \text{ m}^{-3}$, respectively. Both the positions of peaks of the gas temperature and the density of Cu atoms moved quickly from the inside toward the outside of discharge channel; their moving velocities decreased from 1200m/s to 200m/s within 20 μ s.

Key words: Laser scattering techniques, Impulse discharge, Glow-to-arc transition, CARS, LIF, Thomson scattering

1. Introduction

An impulse discharge in low pressure air has a simple discharge channel, its reproducibility and some discharge phases in the discharge process. By this reason, the impulse discharge is convenient to clarify transitional mechanism between discharge phases and characteristics of each discharge phase.^{(1) (2)} Studying the behavior in the discharge process consisting of glow and arc as sparkover phase, it will be also useful to obtain conditions of a stable glow discharge such as an excitation source of discharge pumped gas lasers.⁽³⁾

On the other hand, we have proposed a method for depositing superconductive thin film on a substrate using repetitive impulse discharges.⁽⁴⁾ In this method, however, effects of the impulse discharge on emission and deposition, and characteristics of the species emitted due to the discharges are not yet enough clarified. Therefore, they have to be probed continuously and in detail in order to establish the thin film deposition method.

In that connection, the authors already measured the axial distribution of gas temperature and its temporal variation in the impulse discharge.⁽⁵⁾ Both the relative intensity distribution and the line intensity ratio of rotational spectra of the 2nd positive (0, 1) band of N_2 were used. In these methods for measurement of gas temperature, however, the measurable region is limited to the discharge

channel by reason of depending on the optical emission from the channel.

So, this study aims to reveal the gas dynamic phenomena with a high accuracy in a wide gap space extended to the outside of the discharge channel by using laser scattering techniques. The spatiotemporal distributions of gas temperature were measured using multiplex coherent anti-Stokes Raman spectroscopy (CARS), the electron density using Thomson scattering, and the density of Cu atoms from the electrodes using laser induced fluorescence (LIF). These measurements provide the local values with a spatial resolution less than 0.1mm determined by the laser beam divergence, and a temporal resolution of several ns determined by the laser pulse width.

2. Observation of optical emission process

Figure 1 shows the circuit for generating impulse discharge and the measurement system of optical emission process and electric signals. The needle to plate electrodes having a protrusion on the plate were used here. The gap length between the tips of the needle and protrusion was 20mm, and the discharge chamber was filled with room air of 66.7hPa in pressure.

A positive impulse voltage of 4.0kV (1/300 μ s) was

applied to the needle electrode, which generated the impulse glow-arc discharge. The framing photograph of the gas discharge was recorded by means of an image converter camera, and this was used for identification of the discharge processes and for discussion of the behaviors of the discharge channel. Along with observation of the optical emission process of the gas discharge, the voltage over the gap and the discharge current during the impulse discharge were measured by using a digital oscilloscope.

Fig.1. Measurement system of optical emission

Figure 2 shows (a) the framing photographs, (b) the sketches of them and (c) the waveforms of applied voltage and discharge current of an impulse discharge which turns to arc from transitional glow. In the sketches, the bright regions in the framing photographs are represented by dense dots, and dark regions by sparse dots.

From these framing photographs and the current waveform, it is found that the instantaneous value of current increases as two constricted parts of the positive column (CPCs) extend, and rises rapidly with the occurrence of arc when the glow to arc transition appears at $1.8 \mu\text{s}$ with the positive column still having a diffused part (DPC). The arc is initiated by the establishment of a cathode spot. Also, it seems that the constriction in the glow stage causes the increases of current density and heating of neutrals, and affects the transition to arc. The discharge current reaches a peak value of about 110A at about $2.3 \mu\text{s}$ when two CPCs combine at the middle of the gap.

Fig.2. (a)Framing photographs, (b)sketches of framing photographs and (c)waveforms of discharge voltage and current

3. Measurements of discharge dynamics using laser scattering

3.1 Multiplex CARS for the gas temperature measurement

CARS is one of non-linear Raman scattering techniques characterized by the coherent generation of Raman radiation based on a third order non-linear susceptibility.⁽⁶⁾ CARS consists of the excitation of a Raman-active molecular vibration medium by a pair of high power laser beams. When the difference between angular frequencies ω_1 and ω_2 of the polarized laser beams is close to a Raman active angular frequency ω_r of molecules, the molecular polarization with a frequency of $\omega_3 = 2\omega_1 - \omega_2$ builds up collectively.

Multiplex CARS which uses the beam of ω_2 with a wider linewidth than that of ω_1 can obtain the broad-band CARS spectra. As multiplex CARS can obtain the CARS spectra by only one laser shot, this technique suits the measurement of gas temperature in a transient phenomenon like an impulse discharge.

The gas temperature is determined from comparing the observed CARS spectra with the simulated ones which are

calculated by considering the third order non-linear susceptibility.⁽⁷⁾ The third order non-linear susceptibility is calculated by the spectral profiles of laser beams, the rotational and vibrational structure in nitrogen molecules, and the distribution function. In this study, the intensity distribution of CARS spectra of Q-branch was calculated. By observing the Q-branch CARS spectra of nitrogen molecules in thermal equilibrium, we can obtain a rotational temperature. The rotational temperature is considered to agree with the gas temperature because the relaxation time of transition between the rotational and the translational energies in an air pressure of 66.7hPa is less than 10ns by estimating in consideration of collisional frequency of N_2 and O_2 . Then, the measurement of the order of μs is done in thermal equilibrium. We can determine the gas temperature within an accuracy of $\pm 10\%$ at the room temperature in this technique.

Fig.3. Schematic diagram of multiplex CARS system

Fig.4. Configuration of electrode system and measuring points

Figure 3 shows a schematic diagram of the multiplex CARS system used for observation of the CARS spectra which are Q-branch rotational ones of N_2 (0,1) vibrational band. The ω_1 beam had an energy of 65mJ at 532nm (linewidth 1.0cm^{-1}) of a frequency-doubled Nd:YAG laser. The pulse duration of the laser was about 7ns. The ω_2 was tuned to a wavelength of 607.3nm (linewidth 80cm^{-1})

of a dye laser with an energy of 8mJ pumped by the laser mentioned above. CARS spectra of ω_3 were observed using a spectroscope with an optical multichannel analyzer.

Figure 4 shows a schematic diagram of the configuration of needle to plate electrodes having a protrusion on the plate and the measuring points of the impulse discharge. The gas temperatures were measured at the points of $X=0$ (inside the channel) and $X=2, 4, 6, 8, 10\text{mm}$ (outside the channel) keeping the height constant at each of $Y=1.5, 5, 10, 15, 18.5\text{mm}$ from the tip of the protrusion.

3.2 LIF for the measurement of Cu atom density

The experimental setup for the measurement of Cu density by LIF⁽⁸⁾ is shown in Fig. 5. It consists of a Nd: YAG laser pumped dye laser, a gas discharge chamber and a detection system. In the detection system, a photomultiplier tube is used for sensitive detection.

For the measurement of the density of Cu atoms from the electrodes, a two-level system for laser excitation and fluorescence detection at 324.75nm ($4s^2 S_{1/2} \leftarrow 4p^2 P_{1/2}$) was used. The peak power of the dye laser through a filter was 178W at 324.75nm with a methanol solution of DCM 640 dye. This value was much higher than the saturation power (0.3mW at 324.75nm) calculated in consideration of a spectral width of 0.005nm and a condensed laser cross section of $4.38 \times 10^{-10}\text{m}^2$. The dye laser beam was introduced into a measuring point and focused by a 400mm focal length lens. The LIF signal was observed from the direction normal to the laser beam, through a collecting lens, an optical fiber, a spectroscope and a photomultiplier tube.

As the fluorescence intensity is proportional to the atomic density, the absolute density can be calibrated by the Rayleigh scattering⁽⁹⁾ from a gas with known scattering cross section at room temperature. In this experiment, 66.7hPa of Ar gas was used for that calibration. Here, we neglected the quenching by the collision between Cu atoms and the low pressure air.

Fig.5. Schematic diagram of LIF system

3.3 Thomson scattering for the measurement of electron density

In order to understand the internal mechanisms of discharge plasmas, it is important to clarify the electron behavior because the electrons are initially energized by the discharge heating process and then induce physical and chemical reactions upon other plasma species.

Laser Thomson scattering provides local measurements of electron density and possesses few ambiguities in the interpretation of data.^{(3) (9)} When the scattering parameter⁽¹⁰⁾ is over 1, the scattering is called the "collective scattering". This scattering consists of the contribution of electrons and ions. The spectral width of ion term is much narrower than that of electron term and the scattered power in the electron and ion terms are the same order of magnitude.⁽¹⁰⁾ Then, the measurements were done on the ion term because this scattered emission power per unit wavelength is large.

The experimental setup for the measurement of electron density is almost similar to that of LIF. The second harmonic beam from a Nd:YAG laser, with a wavelength of 532nm, was used as a radiation source for the Thomson scattering experiment. The laser had a pulse energy of 150mJ and a pulse duration of 7ns. Combined with Rayleigh scattering from an Ar gas for the same optical system, the absolute electron density is calibrated within an accuracy of $\pm 15\%$.⁽¹⁰⁾

4. Experimental results and discussions

4.1 Gas temperature, electron density and density of Cu in a discharge channel

The gas temperature and electron density which were obtained near the tips of anode ($X = 0\text{mm}$, $Y = 18.5\text{mm}$) and cathode ($X = 0\text{mm}$, $Y = 1.5\text{mm}$) of the impulse discharge are shown in Figs. 6 and 7. In these figures, the error bars indicate the range of scatter in five different measurements. Both of the gas temperatures obtained near the tips of anode and cathode go up suddenly at the transition from glow to arc, and come up to the highest at the peak of current. The peak values of gas temperature near the tips of anode and cathode are 4800K, 4300K, respectively.

In this stage, the electrons having high density are emitted from the cathode by the growth of arc and the combination of the two proceeding CPCs shown in Fig.2. These phenomena are proved in the variation of the electron density in Fig.7. Both of the electron densities obtained near the tips of anode and cathode behave as same as the variations of gas temperature in the process of initial arc. Therefore, the electrons elevate remarkably the gas temperature near the anode and cathode.

After that discharge process, however, the gas temperature become low, since the stored charge in the electrostatic capacity of the power circuit becomes insufficient. That is, although the gas temperature increases with the discharge current in the beginning, it decreases in keeping with the current, which decreases with the rest of the charge stored in the capacitor. However, the gas temperature near the tip of cathode keeps a little higher than that near the tip of anode since the cathode spot keeps longer than the anode spot.⁽⁶⁾

Fig.6. Gas temperature obtained at near the tips of anode and cathode

Fig.7. Electron density obtained near the tips of anode and cathode

Figure 8 shows the spatiotemporal distribution of gas temperature obtained along the discharge channel. In this figure, the broken line shows the development of the tips of CPCs referred to the results of framing photographs.

The gas temperature of discharge channel is quite low when the discharge phase is diffused glow. However, the gas temperature increases with CPC's proceeding, and of the CPC is higher than that of the DPC at the transitional glow discharge phase. After the transition from glow to arc, both gas temperatures near the anode and cathode are higher than that of the arc column. This phenomenon is probably caused by the higher electron density near the anode and cathode.

Figure 9 shows the variations of Cu atomic density observed near the tips of anode ($X = 0\text{ mm}$, $Y = 18.5\text{ mm}$) and cathode ($X = 0\text{ mm}$, $Y = 1.5\text{ mm}$). From this figure,

it is found that Cu atoms appears even in the transitional glow process. The density of Cu atoms measured near the cathode in this transitional glow stage is higher than that near the anode. The gas temperature near the cathode was also 2700K high at the transitional glow which is higher than the melting point of Cu metal; that of the anode was about 1700K.

Fig.8. Distribution of gas temperature along the gap axis

Fig.9. Variations of Cu atoms density near the tips of the anode and cathode

Accordingly, the measured Cu atomic density before the transition from glow to arc seems to depend on evaporation and sputtering due to the high electric field and the high temperature near the cathode surface. The electric field near the electrodes seems to become higher with the development of CPC and an increase in discharge current. Here, the low ionization potential of Cu will also have an effect on the above result.

In the initial arc process, however, the density of Cu atoms near the anode becomes larger than that near the cathode, and the peak value near the anode reaches about 2 times as high as that near the cathode; whereas the peak gas temperature near the anode was only a little higher than that near the cathode. This phenomenon is believed to be

related to the energy injection mechanism of electrons into the anode or to the ionization / excitation of Cu atoms near the cathode. Further measurements are needed in order to investigate this phenomenon.

4.2 Gas temperature and Cu density near the discharge channel

We measured the spatiotemporal distributions of the gas temperature and the density of Cu atoms at the measuring points of X direction near the anode ($Y = 18.5\text{mm}$) and cathode ($Y = 1.5\text{mm}$) in Fig. 4. It was found that both peaks of the gas temperature and density of Cu atoms at a measuring point outside the channel appear later than those in the channel as shown in Fig. 10.

Figure 11 shows the velocity of individual peaks from the inside to the outside of the channel. It is found that each moving velocity of peaks of gas temperatures and Cu density changes in the same manner⁽¹¹⁾ and moves quickly from the inside toward the outside of the channel; their moving velocities decreased from 1200m/s to 200m/s within 20 μs .

Fig.10. Density of Cu near the anode

Fig.11. Velocities of peak values of gas temperature and Cu density

5. Conclusion

We applied the laser scattering techniques to the diagnostics of the impulse discharge in the low pressure air. The gas temperature was measured using multiplex CARS, electron density using Thomson scattering and the density of Cu atoms from the electrodes using LIF. The results are summarized as follows.

- (1) The gas temperature was higher in the constricted regions, especially at the two parts near the tips of anode and cathode along the positive column.
- (2) The gas temperature obtained near the tips of anode and cathode was higher than melting point of Cu before the transition from glow to arc, and the sputtered and evaporated Cu atoms were measured.
- (3) The gas temperature and the density of Cu atoms obtained near the tips of anode and cathode became high with a rise of discharge current and at the transition from glow to arc, and highest when the discharge current took a peak value as same as the electron density.
- (4) The density of Cu atoms obtained near the tip of anode was about 2 times as high as that near the tip of cathode.
- (5) The spatial evolution of the gas temperatures and Cu density outside the discharge channel had occurred with the same characteristics.

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