NON-EQUILIBRIUM EFFECTS IN PULSE MODULATED INDUCTION THERMAL PLASMA FOR ADVANCED PROCESSING

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An induction thermal plasma system has been newly designed for advanced operation with a pulse modulated mode to control the plasma power in time domain and to create non-equilibrium effects such as fast quenching of the plasma to produce new functional materials in a high rate. The system consists of MOSFET power supply with a maximum power of 50 kW with a frequency of 450 kHz, an induction plasma torch and a vacuum chamber. The pulse modulated plasma was successfully produced at a plasma power of 30 kW and a pressure of 760 torr, with taking the on and off time as 10 and 5 ms, respectively. Measurements were carried out on the time variation of the spectral lines emitted from Ar species. The plasma temperature in an on-off cycle was estimated by the Boltzmann plot method and it changed periodically from 0.5 to 2.5 eV during the cycle as well as deviated considerably from the estimation of thermal equilibrium calculation. Key words: induction thermal plasma, pulse technique, non equilibrium effects, excitation temperature

1. INTRODUCTION

Over the past several decades, a large effort has been devoted to the experimental and numerical analysis of the temperature or the flow fields in the inductively-coupled r.f.(radio frequency) plasma in the steady state, continuos mode. In conjunction with respects to the application of such thermal plasma with a high reactivity to the processing of materials, a special attention has been given recently for the investigation about the dynamic behavior of the induction plasma. Sakuta et al. [1][2] developed first a 1-dimensional model to study the dynamic behavior of r.f. thermal plasma to a sudden change in coil current, and then the 2-dimentional time dependent code has been developed by Mostaghimi et al. [3]. Both calculations showed almost equivalently that the time required to achieve a new steady-state was around 5 to 30 ms for the pressure range from 10 to 100 kPa. This means that if the absent time of the exciting magnetic field is less than the above time constant, the plasma will re-establish again with pulse on action for the electromagnetic field.

This gives an interesting possibility to introduce several important effects in the high-power inductively coupled thermal plasma, that is, 1)Repetitive generation of high and low temperature period, 2)Control of power and heat flux in time domain, 3)Application of extremely high or low electromagnetic field, 4)Introduction of non-equilibrium condition in the electron and heavy particles temperatures as well as in the composition of chemical species including an emphasis of important radicals.

In this paper, an induction thermal plasma system have been newly developed for the advanced material

processing with using a power transistor system with 50-kW power and 450-kHz frequency MOSFET(Metal Oxide Semiconductor Field Effect Transistor). Experimental results are presented for successful operation with pulse modulated mode at a plasma power of 30 kW and a pressure of 760 torr. Dynamic change of the plasma temperature during a pulsing cycle was measured by optical emission spectroscopy and the results showed a strong deviation from the LTE (Local Thermal Equilibrium) calculation. Discussions were made on the comparison of the experimental data with the equilibrium calculation and the total efficiency of the electrical power input to the plasma for the transistor system developed.

2. PULSE MODULATED INDUCTION PLASMA SYSTEM

An induction thermal plasma generation system was developed, which can be operated with both continuous and pulse modulated mode. A new system has a MOSFET inverter power supply which has a rated power of 50 kW at a fundamental frequency of 450 kHz with a high conversion efficiency more than 85%. The induction plasma torch shown in Fig.1 consists of a 10turns coil of 120-mm diameter and 153-mm length, which is considerably large for both radial and axis dimensions compared to the conventional ones associated with the vacuum tube power supply. The main plasma torch has a standard construction of double quartz tubes and the inner one has 70-mm diameter and 370-mm axial length. The transmission of electrical power from the inverter to the plasma is made by a series LC resonance circuit rather than the parallel resonance circuit. This is mainly reflected by the rated

output of 150-V voltage and 460-A current of the inverter. After the successful operation in continuous mode, the pulse mode operation was performed at a power level of 30 kW for Ar-H₂ plasma under atmospheric pressure condition. Fig.2 shows the timedependent pulsing signal installed in the control unit and the modulated magnitude of coil current for the pulse period of 15 ms. The lower level of the current magnitude reaches down to 62% of the maximum, which corresponds to 38% in plasma power level. The rise and fall time of the current magnitude was around several hundreds us. This is much shorter than the inherent time constant of the induction plasma, several ms. Thus, the system developed gives almost ideal pulsing action against the plasma, which is necessary especially for generating the non-equilibrium state in it, or for measuring the dynamic behavior of the plasma.



Fig.1 New Induction Plasma Torch.



Fig.2 Pulse Modulated Coil Current

3. TIME-DEPENDENT PLASMA TEMPERATURE AND CHEMICAL SPECIES

An optical system was used to investigate the plasma condition in spectroscopic mode and the observation position is adjusted at the radial center of the torch and 10mm below from the end of the coil. The light radiated from this position is transmitted to the incident slit of the monochromator (Jobin Yvon HR-320) through a camera lens and an optical fiber bundle. On the output focal plain of the monochromator, the light at two different wavelengths are transmitted to the photomultiplier separately through optical fiber bundles. In this experiment, Ar spectral line at a wavelength of 751 nm and H α line at 656 nm are measured and the data are stored into a personal computer. Another multi-channel system (1024 address) was set up to measure several spectral lines on 100-nm wavelength region at an instance.

The time-dependent characteristics of Ar-H₂ plasma within a cycle of pulse on-off are given in Fig.3 for the plasma power of 30 kW under several pressure conditions. The temperature was estimated from the Boltzmann's plot of several spectral lines emitted from neutral Ar. It can be recognized firstly that the average temperature across the plasma diameter changes periodically from low to high value at any pressure levels, corresponding to the pulsation of the plasma. It should be noticed that such drastic change of the plasma temperature was unexpected from the calculation based on the recent LTE modeling [1][2][5], which will be discussed in the next section. The average temperature across the plasma diameter measured as the exciting temperature of Ar atom shows that it changes periodically from 5,000 to 14,000 K at the middle of the coil, that is in the main discharge region, corresponding to the pulsation of the plasma. Relatively weak change of the excited temperature, on the contrary, can be seen at the downstream portion from 3,000 to 8,000 K, reflecting the weak coupling of the high temperature zone with the electromagnetic field.



Fig.3 Time evolutions of Ar exciting temperature

Another time-dependent radiation intensity of Ar, N, and H spectral lines, which were measured in atmospheric Ar-H₂ and Ar-N₂ pulsing plasmas operated at 17-kW power, is demonstrated in Fig.4. The spectral intensity is normalized here against the steady state level appears around at t=8 to 13 ms after the pulse on t=2 ms in the figure. Each spectral intensity has an inherent rise and fall time constant, for example, N atom has extremely slow rise time, while H atom has a sharp rise and slow decay. This phenomenon cannot be explained again from the LTE modeling and implies an important situation that non-chemical equilibrium effects are occurring in the flux density of N or H radical species corresponding to the pulsing operation. This is supported with a recent theoretical work, where the reaction kinetics of all the chemical species are introduced into the r.f. plasma code [6]. The result shows that after the pulse off of the r.f. coil current, almost 1 to 2 ms is necessary for the plasma concentration to converge to a steady state equilibrium condition in atmospheric N₂ plasma.



Fig.4 Evolution of Ar, H, N spectral intensity



Fig.5 Comparison of Ar line intensity

4. COMPARISON WITH THEORETICAL CALCULATION

Using a 2D-ICP model under LTE assumption, calculations have been done for the same operating conditions. Figure 5 depicts the comparison between experimentally and theoretically obtained spectral intensities. It can be noticed from the figure that the

theoretically predicted intensity responded slowly compared to experimental case after pulse-on. This is due to LTE deviation practically as it has been recognized that plasma remains at non-LTE condition during transition period around lower temperature. Comparison of characteristic delay-times (on-time and off-time) with SCL is illustrated in Fig.6. The delay-time was significantly higher theoretically due to LTE matter, as discussed earlier, but it was almost similar for off-time pulsation because the plasma resides about similar condition for any SCL of same upper-current level just before off-time and thus follows similar trend of response.



Fig.6 Comparison of delay-times

5. EFFICIENCY OF MOSFET INVERTER PLASMA SYSTEM

Fig.7 shows the component of power consumption in induction thermal plasma system for both standard vacuum tube and MOSFET transistor power supply [8]. The power dissipation was measured at each part by calorimetric method using water flow. As a common power consumption part for both system are the discharge tube and the work coil, which are around 13% and 7%, respectively. The most remarkable difference can be found in the power source part, that is, 60% for vacuum tube system and 5% for MOSFET transistor system. The difference reflects directly the input power to the plasma and especially high efficiency of around 70% is achieved in the MOSFET system, while a quite low efficiency of 20% is estimated in the conventional vacuum tube system. This is an example of the measurement of power dissipation for a certain system exists. The actual situation is, however, not so far from the result obtained here.

Fig.8 explains schematically the concept of pulsation in thermal plasmas and this concept has been introduced here firstly. The pulsing technique has been relatively well understood and utilized in the low temperature, cold plasma region, mainly for increasing the energy and the number density of electron and radical species, keeping the escape from thermal breakdown. On the other side, the thermal plasma is established only after such dielectric breakdown of cold gaseous medium and it has a quite steady feature for the excess energy input. The highpressure plasmas in inductively coupled or dc mode as well as the nuclear fusion plasma are typical examples of such steady and final mode of plasmas after cold plasmas. Obviously, a transition phase exists between these two types of plasmas which can be recognized as the breakdown or extinguish phase, if it is viewed from the cold plasma or thermal plasma side, respectively. The introduction of pulsation in the thermal plasma region gives a returning back to the transition phase in its pulse off period, escaping from extinguish or recovery of dielectric strength by pulse on action.



Fig.7 Power dissipation in plasma generation for MOSFET of induction thermal plasma

The main feature of the transition phase is that several non-equilibrium effects are existing, including a difference between electron and heavy particle temperature, radical species density as well as chemical reaction rates, which are deviated considerably from the equilibrium state. This unique phase should be more utilized for the material processing, especially the synthesis of new functional material with high efficiency and high rate. Thus, the meaning of the pulsation in the thermal plasma side is considered to give a quick quenching or cooling of the



Fig.8 Concept of pulsation in cold and thermal plasma

plasma in the time domain, aiming at returning to the transition phase where the several chemical species co-exist under non-equilibrium condition in both its energy and concentration.

6. SUMMARY

The inductively-coupled radio frequency plasma was successfully generated under a pulse modulated mode at a high pressure of 100 kPa and a power of 40 kW, by using semiconductor inverter power supply. Although the power electronics technique gives the standard feasibility to control the plasma power in time domain rather than the conventional amplitude domain, its characteristic feature gives other important possibilities to produce intentionally a non-equilibrium state of particle temperatures and the flux density of radical species or to control the thermal flux to the substrate in case of synthesis or spraying, both aiming at an advanced material processing. A further effort should be paid to the scaling up of the induction thermal plasma technology in high power level of several hundreds kW with a reasonable electrical efficiency, which is requested now from the industrial side especially for high rate processing of destruction of several contaminants for earth circumstance.

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