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Non-equilibrium effects in pulse modulated induction thermal plasma for advanced material processing*

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Abstract: 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 with a 10-turns coil of 80 mm diameter and 150 mm length 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 5 ms, respectively. Measurements were carried out on the time variation of the spectral lines emitted from Ar species. The dynamic behavior of the plasma temperature in an on-off cycle was estimated by the Boltzmann plot method and the excitation temperature of Ar atom changed periodically from around 0.5 to 2.5 eV during the cycle. Non-equilibrium effects are found in this Ar exciting temperature as well as species number of Ar, H and N, in the plasma under pulse mode operation.

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 RF (radio frequency) plasma in the steady state, continuous 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 one-dimensional model to study the dynamic behavior of RF thermal plasma to a sudden change in coil current, and then the two-dimensional 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 be reestablished 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 (i) Repetitive generation of high and low temperature period, (ii) Control of power and heat flux in time domain, (iii) Application of extremely high or low electromagnetic field, (iv) 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.

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In this paper, an induction thermal plasma system has been newly developed for the advanced material processing with introducing the pulse modulation mode, which may create non-equilibrium effects in high pressure thermal plasmas. After explanation of the new 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. Non-equilibrium effects are also found in number of radicals such as Ar, H and N in the plasma under the pulse modulated mode. Discussions were made on the limitation of the stable region of pulse operation, and on the comparison of the efficiency of the power input to the plasma for the developed transistor system and the conventional vacuum tube system.

PRINCIPLE OF PULSE MODULATION

In order to achieve the pulse modulation of the magnitude of the coil current oscillating with a high frequency around MHz, the solid state transistor devices are necessary to be introduced newly, rather than the conventional vacuum tube oscillator technique. Figure 1 shows the applicable region of the semiconductor inverter power supply in the power and the frequency, which has been developed and extended in the power electronics region. Among several high power transistor device appears in the commercial stage, MOSFET (Metal Oxide Semiconductor Field Effect Transistor) or SIT (Static Induction Transistor) is most promised candidate for our trial of generating the pulse-modulated induction thermal plasma because the applicable range of the frequency high as 500 kHz, which is within the range capable of exciting the gaseous medium in a conductive plasma state. Figure 2 indicates a comparison of two coil currents in normal continuous mode and in pulse modulated mode. In the normal mode shown in Fig. 1a, the amplitude of the coil current is constant and the characteristic time constant of the thermal plasma itself, ms order, is much larger than the period of 2 μs which corresponds to 450 kHz, an inductively coupled plasma can be established under a steady mode. In the case of Fig. 1b, where the pulse modulation is operated onto the amplitude of the coil current with a cycle of 10 ms order. In this case, corresponding to the pulse on, off action of the magnetic field, the plasma should have a transient components, because the frequency of the disturbance is as low as the plasma can follow it [2,3]. By controlling the duty factor and the current ratio in the pulse on and off period, new feasibility and effects are expected to be added to the high-power thermal plasmas as mentioned above. The principle limitation of such pulse modulation of the induction thermal plasma was recognized already in both the theory and experiment, in terms of maximum pulse off time of around 10 ms, beyond which the plasma diminishes due to lack of the electrical conductivity [4].

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. 3 consists of a 10-turns 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 40 kW for Ar-H_2 plasma under atmospheric pressure condition. Figure 4 shows the time-dependent 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 40% of the maximum, which corresponds to 16% in plasma power level. The rise and fall time of the current magnitude was around several hundred microseconds. This is much shorter than the inherent time constant of the induction plasma, several milliseconds. Thus, the system developed gives almost ideal

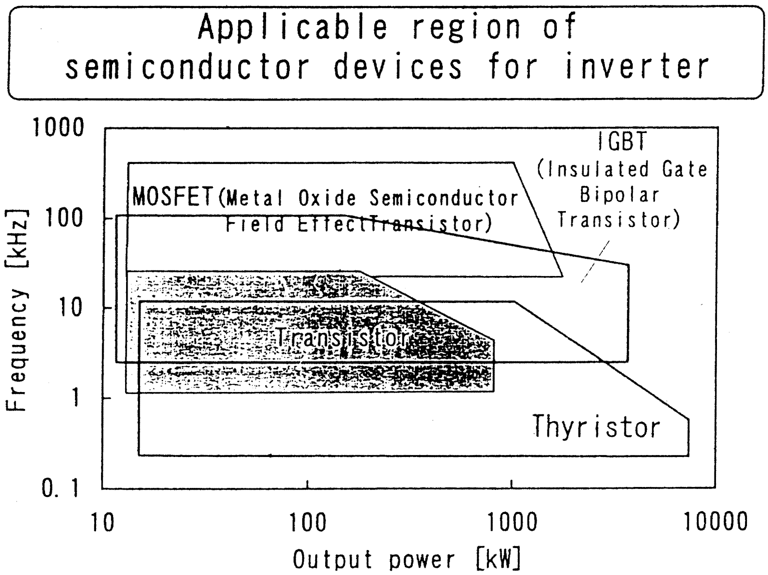

Fig. 1 Applicable region of power transistor.

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.

TIME-DEPENDENT PLASMA TEMPERATURE AND CHEMICAL SPECIES

Figure 5 shows an optical system used in the experiment. Observation position is adjusted at the radial center of the torch and 10 mm 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, as shown on the right hand side of Fig. 3. On the output focal plain of the monochromator, the light at two different wavelengths are transmitted to the photomultipliers separately through optical fiber bundles. In this experiment, Ar spectral line at a wavelength of 751 nm and Hα line at 656 nm are measured and the data are stored into a personal computer. On the left hand side in Fig. 3, 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-H_2 plasma within a cycle of pulse on-off are given in Fig. 6 for the plasma power of 11 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

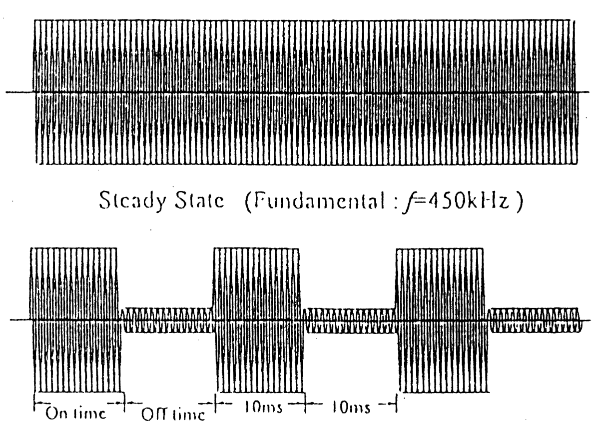

Fig. 2 Principle of pulse modulating.

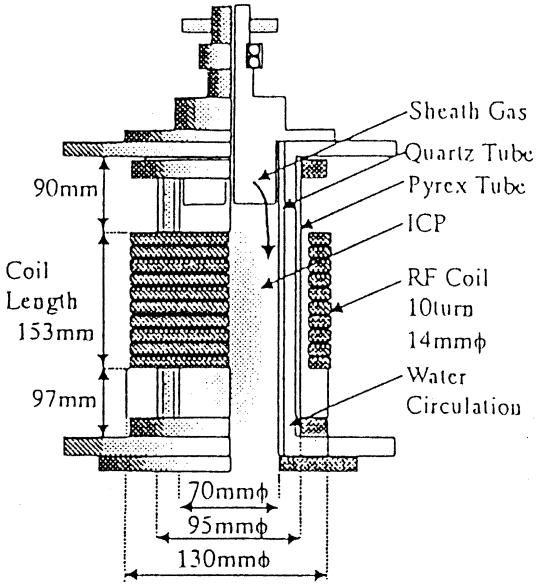

Fig. 3 New induction plasma torch.

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] and the experiment shows a large deviation from the equilibrium condition of electron and heavy particle temperature even under the atmospheric pressure level [5]. Figure 6, (a) and (b) show the pulsing signal and the time-dependent characteristics of the spectral line intensity obtained in a pulse modulated induction plasma at 11 kW power for different pressures of 27, 53 and 101 kPa, with taking the repetitive period of 5 ms and the duty factor of 50%. It is found that the plasma is successively produced in repetitive mode rigidly corresponding to the pulsing signal. The emission spectrum of Ar 751 nm as well as the continuum radiation change between high and low levels with rise and fall time constant around several ms order as has been numerically estimated previously [4,5]. It should be noticed, from this figure, that the radiation intensities of the Ar line and the continuous spectra increases with the pressure as has been expected from theoretical analysis.

The time-varying plasma temperature at different pressures during the pulse on-off period is given in Fig. 6c, which was obtained from the Boltzmann plot of five spectral lines emitted from Ar Neutral lines.

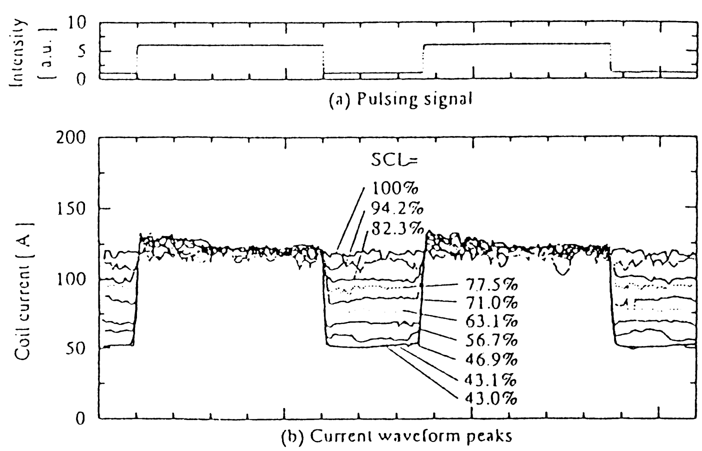

Fig. 4 Pulse modulated coil current.

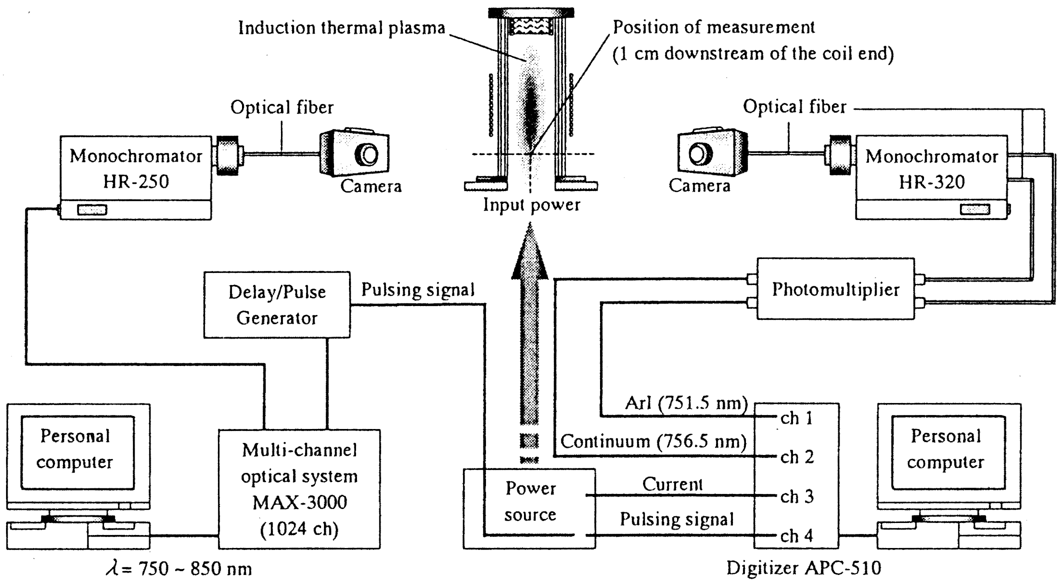

Fig. 5 Optical measurement system.

The average temperature across the plasma diameter measured as the exciting temperature of Ar atom shows that it changes periodically from 5000 to 14 000 K corresponding to the pulsation of the plasma. Furthermore, the temperature changes more strongly with decreasing the pressure, since the specific heat for low pressure plasma is smaller than that for high pressure one.

Another time-dependent radiation intensity of Ar, N and H spectral lines which were measured in atmospheric Ar-H_2 and Ar-N_2 pulsing plasmas operated at 17 kW power is demonstrated in Fig. 7. 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 phenomena 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

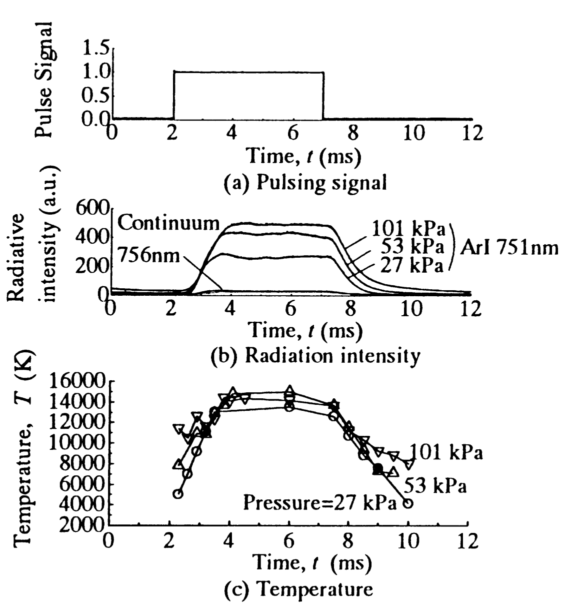

Fig. 6 Time evolutions of Ar exciting temperature.

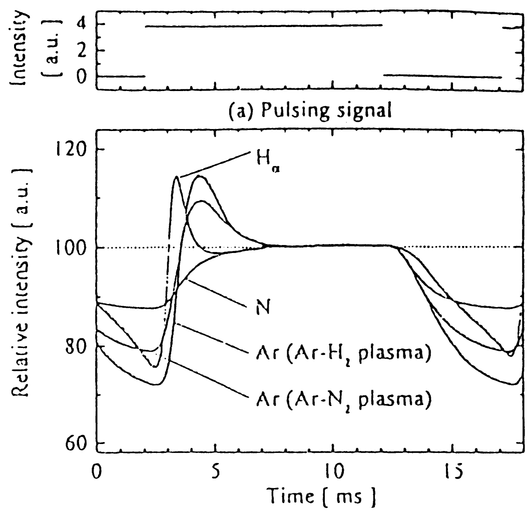

Fig. 7 Time evolution of emissions from Ar, N and H.

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 RF plasma code [6]. The result shows that after the pulse off of the RF coil current, almost 1 to 2 ms is necessary for the plasma concentration to converge to a steady state equilibrium condition in atmospheric N_2 plasma.

OPERATION REGION AND EFFICIENCY OF PULSED INDUCTION PLASMA

Figure 8 shows the range of limitation of the pulse operation obtained in the experiment under a fixed condition of 17 kW input power and 101 kPa pressure for the induction torch developed here. The bold lines indicates the maximum off-time for sustaining the pulse modulated plasma at different on-times, which were obtained several experiments. The dotted line as an approximate straight line indicates, therefore, the limit of the stable operation region only under which the pulsing of the induction plasma is possible. In the case of the shimmer current level (SCL) of 60%, the maximum off time T_{off} for the pulsation is approximately constant value of 10 ms when the pulse on time T_{on} is larger than 5 ms. The results clearly indicates that the time necessary for the plasma to recovery fully to the previous continuous

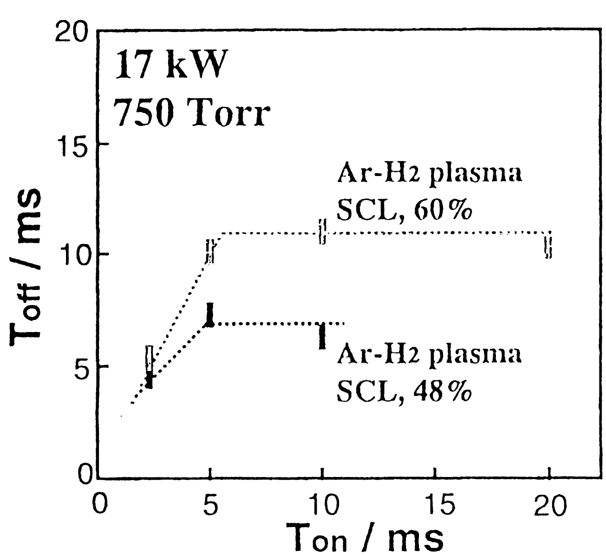

Fig. 8 Limitation of the pulse modulated operation of induction thermal plasma.

level is at least 5 ms. In the case of shorter on time T_{on} less than 5 ms, the maximum pulse off time depends on the T_{on} reflecting that the joule input is not sufficient for full recovery of the plasma. Similar tendency is recognized also in the case of SCL = 0.48. In this case, the maximum T_{off} becomes relatively shorter around 6 to 7 ms, compared with 10 ms in SCL = 0.6 case. This value agrees with the theoretical one estimated from the mathematical model above mentioned.

Figure 9 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 systems 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 reflect 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 systems exist. The actual situation is, however, not so far from the results obtained here.

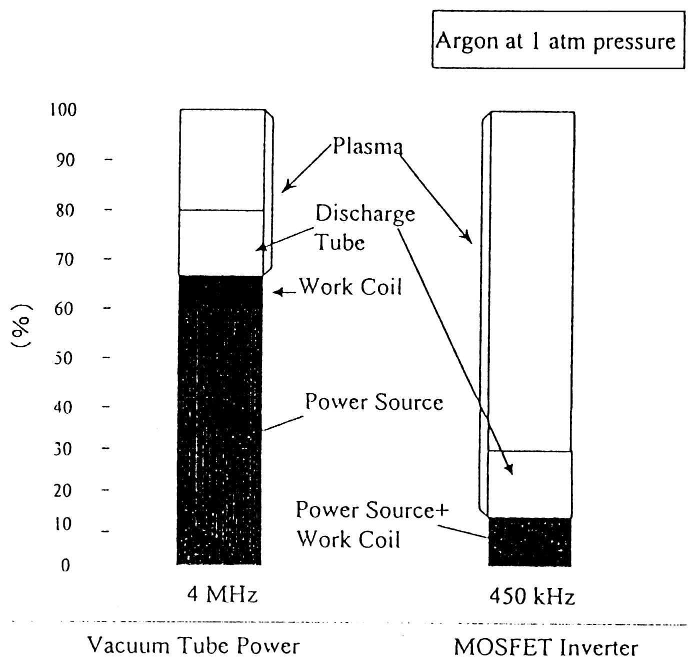

Fig. 9 Power dissipation in plasma generation for MOSFET transistor system (after Sakakibara *et al.* [8]).

Figure 10 explains schematically the concept of pulsation in thermal plasmas which has been introduced firstly here. 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 high pressure 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.

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

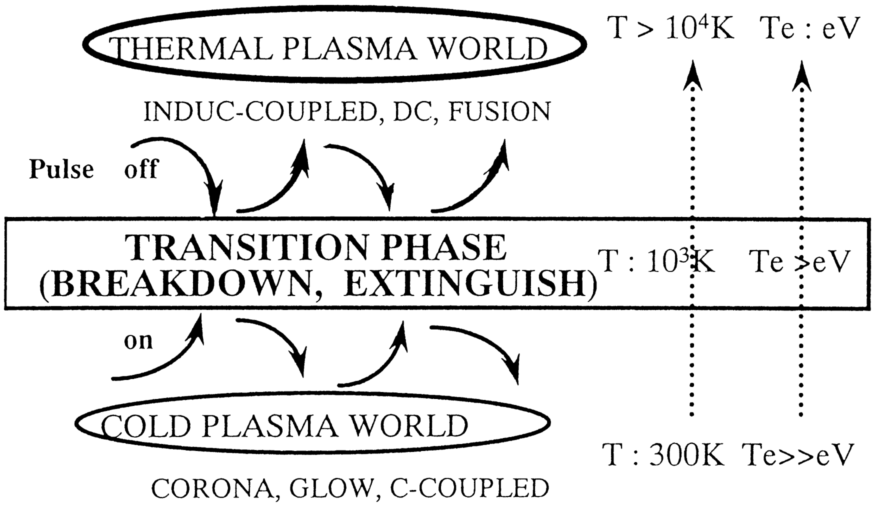

Fig. 10 Concept of pulsation in cold and thermal plasmas.

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 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.

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 hundred 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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