

# The Effect of Excitation Frequency ( $f=10$ kHz-100 MHz) on the Formation of Excited Atoms and Radicals

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## Abstract

The formation of excited argon atoms and radicals from nitrogen-hydrogen plasma were carried out using two kinds of plasma reactors, the excitation frequency of which can be changed between 10 kHz and 100 MHz. Optical emission spectroscopy and probe technique were applied to identify the species and to estimate electron temperature and electron density, respectively. The results obtained by varying frequency are discussed, together with those obtained by changing pressure and power input. Moreover, the net power to create plasma was estimated by measuring  $V_{pp}$ ,  $I_{pp}$ , and phase change.

## Introduction

It is well known that many parameters influence a given plasma process [1]. Among these parameters, pressure, power input, and excitation frequency are representatives. Although pressure and power input can be easily changed, there are many problems in changing frequency. There are some recent publications on the effect of excitation frequency on plasma processing [1-3]. The frequency effect is explained in the terms of  $\nu/\omega$ . The electron density increases with increasing frequency. In many cases, the region for the changing frequency is narrow, the frequency is not continuous, or the power input is too low to use. This paper describes the results obtained from plasma diagnostics for Ar and  $N_2$ - $H_2$  plasmas formed in high-frequency ( $f=10$  kHz-100 MHz) discharges using two types of plasma creators. The influence of excitation

frequency is discussed together with the results when the pressure and power input were changed. Moreover, the net power,  $P_{NET}$ , to create plasma was estimated by measuring  $V_{pp}$ ,  $I_{pp}$ , and phase shift.

## Experimental

The experimental setups are shown in Fig. 1. Pure Ar and  $N_2$ - $H_2$  (25 vol %) mixture were used as plasma gases. Ar is usually used in order to estimate the condition of plasma and  $N_2$ - $H_2$  is the simplest system including the gas phase reaction. The plasma was created using a capacitive coupled and inductive coupled high-frequency discharges. When capacitive coupled discharge was used, the distance between electrodes was kept constant at 30 mm. For inductive coupled discharge, fused quartz discharge tube with a diameter of 20 mm and a length of 1m [4]. Several kinds of matching networks were used for the plasma creation. Optical emission spectroscopy (OES) was carried out in order to identify the species and the transitions in the plasmas. In order to avoid to cause plasma to fluctuate, a triple probe technique was used to measure electron temperature ( $T_e$ ) and electron density ( $n_e$ ).  $V_{pp}$ ,  $I_{pp}$ ,  $V_{DC}$ , and phase shift were measured between matching box and plasma creator.  $P_{NET}$  was calculated using following equation;

$$P_{NET} = (V_{pp} \times I_{pp} / 8) \times \cos\phi$$

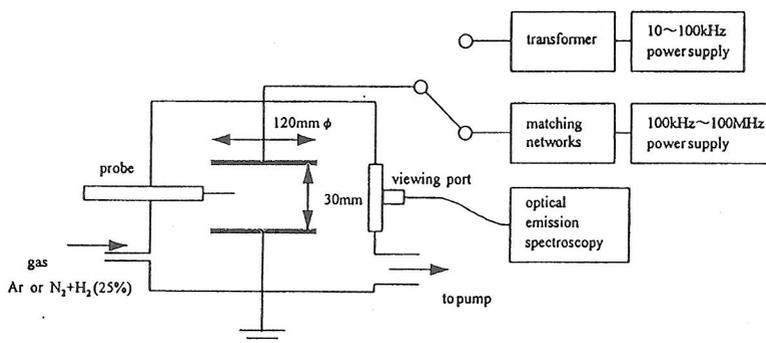


Fig. 1 Experimental setup

## Results and Discussion

Figures 2 and 3 show the dependence of electron temperature,  $T_e$  and electron density,  $n_e$  on excitation frequency in Ar and  $N_2$ - $H_2$  plasmas, respectively. In these cases, pressure was kept at 40 Pa and power input at 100 W. The probe was placed at

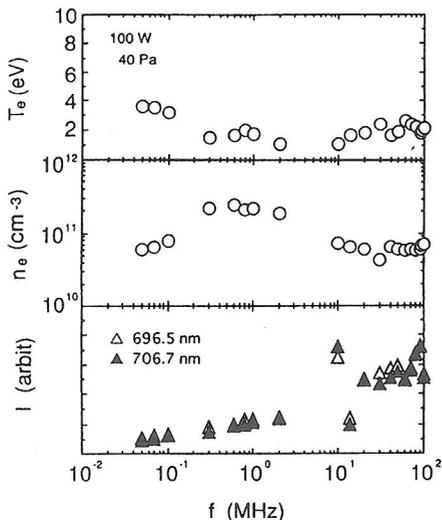


Fig. 2 Effect of frequency (Ar)

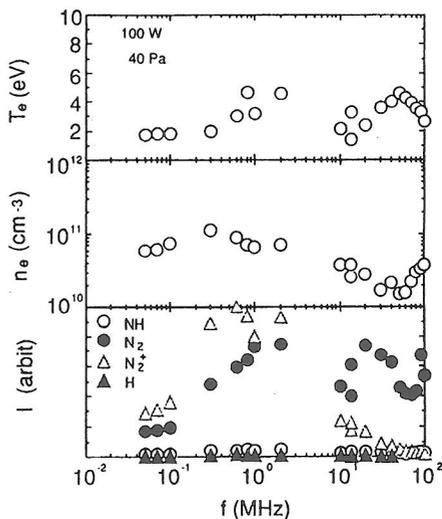


Fig. 3 Effect of frequency (N<sub>2</sub>-H<sub>2</sub>)

the center of electrodes. When the frequency was varied in both plasmas,  $T_e$  continued to remain approximately constant. Meanwhile,  $n_e$  attained a maximum value around a few hundred kHz. OES was carried out to compare these values with intensities of atomic lines and spectra due to emissive species. An optical fiber scope was mounted in the reactor, such that it gazed at the center of the plasma. This was approximately the same point where the probe measurement was made. The changes in intensities of atomic lines in the Ar plasma ( $\lambda=696.5$  nm and 706.7 nm, which correspond to  $3^3P-3^1P^0$  transition) are also shown in Fig. 2. The intensities due to Ar atomic lines increased with increasing excitation frequency. Although the changes in intensities of atomic lines corresponded to the electron density in Ar plasma when pressure and power input changed, the changes did not correspond to the densities in this case. As shown in Fig. 3, the changes in intensities in N<sub>2</sub>-H<sub>2</sub> plasma were different from those in Ar plasma. In this case, the emission peaks due to N<sub>2</sub> ( $C^3\Pi_g-B^3\Sigma_u^+$ , 0-0), NH ( $A^3\Pi-X^3\Sigma^-$ , 0-0), N<sub>2</sub><sup>+</sup> ( $B^2\Sigma_u^+-X^2\Sigma_g^+$ , 0-0), and H(H $\beta$ ) were selected for the comparison of intensity. The intensities of emission spectra due to NH, N<sub>2</sub><sup>+</sup>, and H, which are important species for the formation of nitrogen compounds [4], had a maximum at around a few hundred kHz. This tendency corresponded to the change in electron density. It is suggested that large amount of ions and electrons were formed in the plasma which was created at around a few hundred kHz. While, the intensity of the emission spectrum due to N<sub>2</sub>

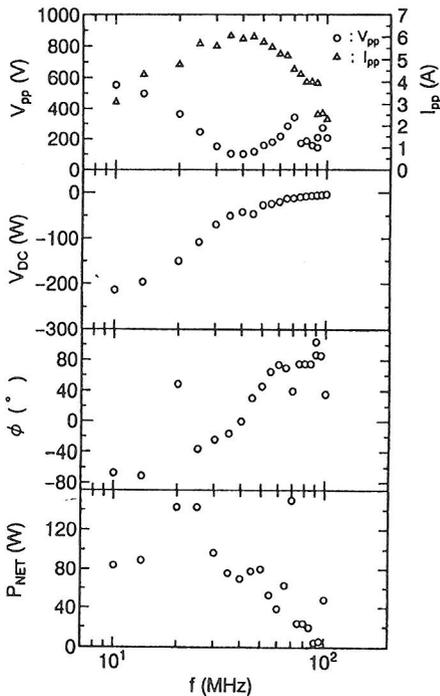


Fig. 4 Effect of frequency (Ar)

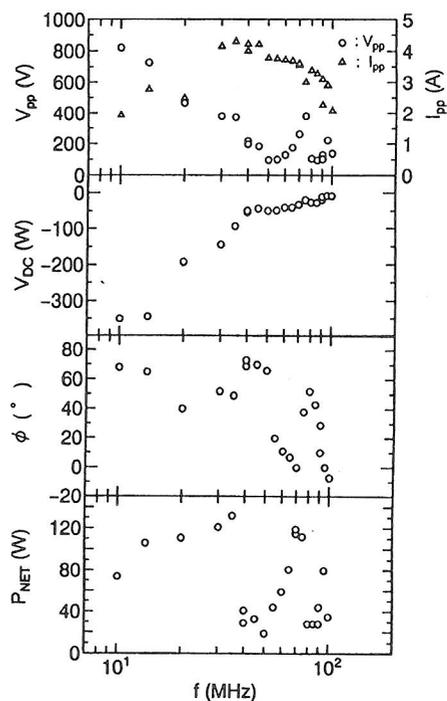


Fig. 5 Effect of frequency (N<sub>2</sub>-H<sub>2</sub>)

increased with increasing excitation frequency, when less than 1 MHz and became saturated over 1 MHz. Although the excitation frequency influenced the electron density, excitation of atoms and formation of radicals, changes in pressure and power input were more dominant in influencing these factors. In both plasma,  $V_{DC}$  gradually increased with increasing excitation frequency. When the excitation frequency was changed,  $P_{NET}$  also should be considered. The results for both plasma are also shown in Fig. 4 and 5. For Ar plasma, the phase shift showed that the load including the plasma played a role as a capacitor, when the frequency was less than 50 MHz. The load played as an inductive coil more than 50 MHz. However, the load played a role as a capacitor for N<sub>2</sub>-H<sub>2</sub> plasma.  $P_{NET}$  for Ar plasma was lower than 30 % of input power and had a maximum at around 20 MHz. Although this tendency was approximately the same to that for N<sub>2</sub>-H<sub>2</sub> plasma less than 10 MHz, the changing tendency was different from that for Ar plasma more than 10 MHz.  $P_{NET}$  had two peaks at around 30 MHz and 70 MHz in the case of N<sub>2</sub>-H<sub>2</sub> plasma. By recalculating the electron density using this  $P_{NET}$  for both plasmas, the values of electron density

seemed increase with increasing excitation frequency. The electron density at 100 MHz could be estimated to be one order of magnitude higher than that at 10 MHz. Since the obtained results can be reconsidered with factors of the  $P_{NET}$ , drastic change in the values will be obtained as predicted. However, the dramatic change of electron density did not occur with changing excitation frequency although the change would be gradual increase with increasing frequency. Moreover, these parameters for  $N_2-H_2$  plasma were measured with changing excitation frequency at 4 Pa. Although approximately the same tendency to that carried out at 40 Pa, the tendency of the change in phase shift,  $\Phi$  was slightly different. As detected in case of Ar plasma at 40 Pa, the load played as inductive coil more than 50 MHz. This results showed the difficulty of the generation of plasma more than 50 MHz at lower pressure. Moreover,  $N_2-H_2$  plasma was formed using an inductive coupled plasma reactor at 40 Pa with changing frequency. In this case, the power input was kept at 50 W. Increase of the emission intensities and decrease of  $V_{pp}$  and  $I_{pp}$  were observed with increasing excitation frequency within 10 MHz-30 MHz. Moreover, it is found that it is difficult to create plasma more than 30 MHz by this technique.

$T_e$  and  $n_e$  for Ar plasma created by capacitive coupled discharge at 13.56 MHz as functions of pressure and power input are summarized in Figs. 6 and 7, respectively. For  $N_2-H_2$  plasma, approximately the same tendency to that for Ar plasma was obtained.  $T_e$  decreased and  $n_e$  increased with increasing power input. However, when pressure was changed,  $T_e$  had a maximum and  $n_e$  had a minimum at approximately 40 Pa. The intensities of Ar atomic lines increased with increasing power input and pressure as shown in Figs. 6 and 7. When pressure was changed,  $P_{NET}$  gradually decreased with increasing pressure as shown in Fig. 6. When power input was changed,  $P_{NET}$  increased and  $V_{DC}$  decreased with increasing power input.

## Conclusion

Excitation frequency influenced the electron density, excitation of atoms, and formation of radicals, however, pressure and power input had a greater influence of these factors. Changes in emission intensities were approximately the same as those in electron density for both plasmas, though monotonous change of electron density with changing frequency could not be found. From our results, it is possible to say that the changing excitation frequency would be effective to capacitive coupled plasma rather than inductive coupled plasma. The phenomena occurred by changing frequency was slightly different depending on the plasma gas.

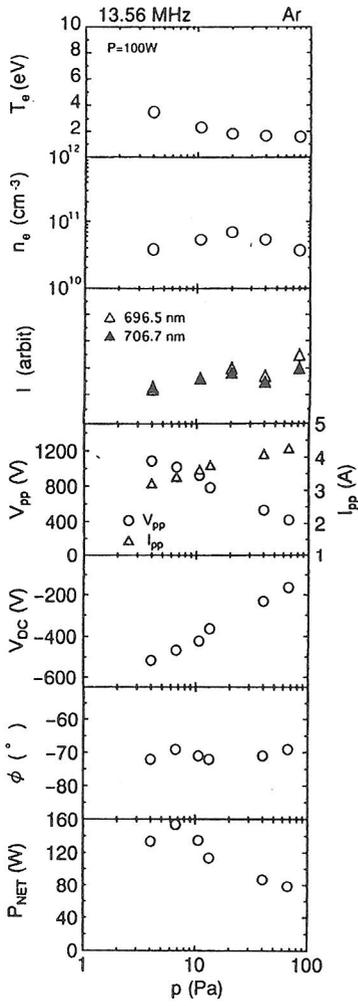


Fig. 6 Effect of pressure

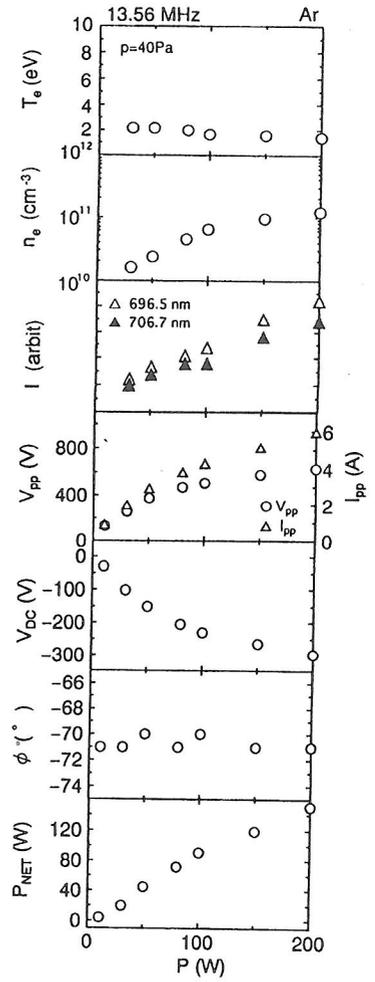


Fig. 7 Effect of power input

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