Nitrogen Atom Density Measurements in NAGDIS-T Using Vacuum Ultraviolet Absorption Spectroscopy

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For the development of a high nitrogen atom density source, we produced nitrogen plasmas and measured the nitrogen atom density using vacuum ultraviolet absorption spectroscopy (VUV AS) at high neutral gas pressure (>1.5 mTorr) and discharge power (>500 W) in the NAGDIS-T, which could generate spiral-shaped dissociative recombining plasmas. It was seen that the nitrogen atom density increases with increasing neutral gas pressure and discharge power, and it reached 6.2 × 10^{17} m^{-3}. In the low gas pressure case, the estimated atom density was confirmed to be consistent with that measured by the actinometry method.

Keywords: nitrogen atom, plasma, NAGDIS-T, vacuum ultraviolet absorption spectroscopy, actinometry

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Nitrogen atoms (N) are expected to be used in various applications such as NOx removal [1], material nitriding, and the molecular beam epitaxy method [2]. A high nitrogen atom density (n_N) environment is required; however, it is difficult to achieve due to the high binding energy of nitrogen molecules (8.80 eV) [3]. For the development of a high n_N source, a torus-type plasma generator NAGDIS-T [4] is used in this study. This device aims to create a high n_N environment through dissociative recombination by generating a spiral plasma with a long connection length of magnetic field lines, which realized high-density nitrogen molecule plasma (N_2^+) generation in the upstream region and its temperature reduction before reaching the wall [5].

In previous studies using the NAGDIS-T, n_N in the ground state was measured spectroscopically by assuming coronal equilibrium [5] and by actinometry [6]. As a result, it was confirmed that n_N was on the order of 10^{17} m^{-3}. In this study, we performed vacuum ultraviolet absorption spectroscopy (VUVAS) to measure n_N in a more direct manner. In Ref. [6], VUVAS was used to compare with actinometry under limited experimental conditions; the present study extends the conditions to higher neutral pressure, p_n, and higher discharge current, I_d, to generate a higher n_N.

Figure 1(a) shows a schematic diagram of VUVAS and actinometry measurement systems in the NAGDIS-T. A vertical and toroidal field of 2.7 and 40 mT, respectively, are applied to create a spiral plasma that is generated from the top inside the vacuum vessel. Because the plasma ro-

![Fig. 1 (a) Schematic diagram of VUVAS and actinometry measurement systems. (b) PMT signals without (blue) and with (red) the nitrogen plasma discharge.](image-url)
O2, N2, and H2 gases for the discharge, is used for the light source [7], and the transmitted light is detected by a VUV spectrometer (NU-Rei Co. Ltd.) and a photomultiplier tube (PMT). VUVAS can detect N atoms that reach around the 2nd loop of the plasma, while the actinometry measures the 1st loop of the plasma. The light emission at the center of the plasma is collected through a lens and transferred to a spectrometer via an optical fiber.

Figure 1 (b) shows typical time evolutions of the transmitted light intensities at 120.0 - 120.1 nm, which corresponds to an N atomic transition, with and without the nitrogen plasma. Here, background nitrogen gas pressures are the same. The light emission from the MHCL is generated in pulses. It is confirmed that the PMT signal decreases when the N plasma is generated, indicating that there are N atoms absorbing the light emission from the MHCL. In addition, we confirmed that no light absorption occurred for oxygen (130.2 nm) and hydrogen (121.6 nm) emissions. For actinometry, 1% Ar is introduced into the nitrogen plasma, and an N emission line at $\lambda = 746.8$ nm and an Ar emission line at $\lambda = 750.4$ nm are measured. Then, $n_N$ is calculated from the ratio of the line emission intensities [6].

Figure 2 (a) shows $p_n$ dependences of $n_N$ measured with VUVAS at $I_d = 5$ and 10 A and actinometry at $I_d = 5$ A. In the VUVAS measurement, the same assumptions were made as in Ref. [8]; i.e., the gas temperatures used in the measured medium and the MHCL were 300 and 400 K, respectively. Here, it should be noted that VUVAS and actinometry measurements at $I_d = 5$ A were not done simultaneously. Although these measurements were performed with the same discharge current, the discharge voltage was not the same, mainly due to some differences in the surface conditions on discharge electrodes and the wall in NAGDIS-T. Figure 2 (b) shows discharge powers ($W_d$) as a function of $p_n$ in corresponding three discharge series. It is seen that $W_d$ decreases as $p_n$ increases, because the discharge voltage decreases with $p_n$. Moreover, $W_d$ in the actinometry measuring discharges at lower $p_n$ condition are slightly higher than those during the VUVAS measurement.

In Fig. 2 (a), it is seen that $n_N$ from VUVAS increases with $p_n$. Also, the $n_N$ at $I_d = 10$ A are roughly two times higher than those at 5 A. At $p_n = 4.74$ mTorr and $I_d = 10$ A, $n_N$ reached $6.2 \times 10^{17}$ m$^{-3}$. The actinometry resulted in nearly consistent values with the VUVAS and it decreases with $p_n$ [6].

To considering the $p_n$ effect on the nitrogen atom generation, $n_N$ normalized to $W_d$ was plotted as a function of $p_n$ in Fig. 2 (c). It is found that values of $n_N/W_d$ at a specific $p_n$ are nearly the same in the three different discharge conditions. Further, monotonical increases with $p_n$ are confirmed.

It is thought that increasing $p_n$ and $W_d$ will lead to the creation of a high $n_N$ environment. An increase of $W_d$ contributes to the increase in $n_N$ because high $W_d$ should increase the electron density, which will promote dissociative recombination and direct dissociation. An increase in $p_n$ promotes a decrease in the electron temperature, causing an increase in $n_N$. The production rate of N via direct dissociation is $\sim 1.6$ times larger than that of dissociative recombination at $p_n = \sim 0.6$ mTorr when using the parameters measured in the 1st loop of the plasma. In contrast, at $p_n \sim 1.5$ mTorr, the production rate of dissociative recombination is $\sim 7.6$ times larger than that of direct dissociation, primarily because the high energy component of the electron energy distribution function decreases. The contribution of dissociative recombination likely increases with increasing $p_n$, mainly due to the decrease in electron temperature.

In this study, we succeeded in measuring the $n_N$ at high $p_n$ (>1.5 mTorr) and high $W_d$ (>500 W) using VUVAS and confirmed that $n_N$ was $2.5 - 6.2 \times 10^{17}$ m$^{-3}$. It was also confirmed that $n_N$ increases with increasing $p_n$ and $W_d$. This is because an increase in $p_n$ causes a decrease in electron temperature, and an increase in $W_d$ causes an increase in electron density, which promotes dissociative recombination and direct dissociation. It would be interesting to further investigate to what degree $n_N$ can be increased by
increasing $p_n$ and $W_d$.

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