Preliminary Experiments of Cavity Enhanced Absorption Spectroscopy for Plasma Torch Flow Diagnostics

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Cavity enhanced absorption spectroscopy system using a diode laser was applied to the plasma flow diagnostics. An absorption line of ArI 842.46nm in a plasma torch flow was targeted and the sensitivity enhancement was evaluated. As a result, the sensitivity was successfully increased by two orders of magnitude compared with the conventional laser absorption spectroscopy.

Nomenclature

Α	=	Einstein coefficient
С	=	velocity of light
d_0	=	absorption length
$d_{ m eff}$	=	effective absorption length
8	=	statistical weight
i	=	absorbing state
j	=	excited state
Ι	=	transmitted laser intensity
I_0	=	probe laser intensity
k	=	Boltzmann constant
Κ	=	integrated absorption coefficient
k_{ν}	=	absorption coefficient
M_A	=	atomic mass
n	=	number of reflections
n_i	=	number density of absorbing state
R	=	reflectivity of mirrors
Т	=	transmittance of mirrors
$T_{ m tr}$	=	translational temperature
ν	=	laser frequency
ν_0	=	center absorption frequency
$\Delta v_{\rm D}$	=	full width at half maximum of Doppler profile
$\Delta v_{\rm L}$	=	full width at half maximum of Lorentz profile

I. Introduction

In past five decades, high enthalpy flow generators have been developed to simulate re-entry conditions for testing the thermal protection system(TPS).¹ However, their exact plume conditions are mostly unknown because they are usually in strong thermo-chemical non-equilibrium. Therefore, measurement of their chemical composition and enthalpy is useful for the evaluation of TPS, and also for the validation of CFD models of non-equilibrium flow.

In our previous study, laser absorption spectroscopy (LAS) has been applied to plumes of arc-heaters and inductively coupled plasma generators^{2,3} to measure the number density of meta-stable OI and translational

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temperature. As a result, strong absorptions of OI 777.19nm line were observed in oxygen and argon/oxygen flows. However, the absorption line of OI could not be detected in a nitrogen/oxygen flow even in a shock layer formed in front of test materials as well as in a plume, because the fractional absorption of OI 777.19nm line in nitrogen/oxygen flow was less than our measurement limit of 1%.⁴ In other group, it was also reported by S. Kim *et al.* that in the NASA Ames IHF (60MW) arcjet facility, the laser absorption was not detected in the plume of nitrogen/oxygen flows, although it could be detected inside the arc heater⁵. To detect the OI absorption in such nitrogen/oxygen flows, sensitivity of LAS have to be enhanced by two orders of magnitude.

Recently, cavity ring down spectroscopy (CRDS) has been employed as a high sensitive laser absorption spectroscopy.^{6,7} The detection limits of the number density for many species have been demonstrated to be from $10^{13}-10^{16}$ [m⁻³].⁸ A ringdown cavity is composed of two concave high reflectivity mirrors, which are aligned at a distance such that stable optical resonator is obtained. Laser light is pulsated below μ s by acoustooptic modulator⁹ or mechanical chopper,¹⁰ and the decay rate of a photon population in an optical cavity is used to obtain the associated total intra-cavity losses (per pass) as a function of the photon frequency. However, because of the weak transmitted laser power of the order of nW, a very sensitive detector is needed.

In conventional LAS, a diode laser is often used for its low cost and easiness of the wavelength modulation. Therefore, in this study, Cavity Enhanced Absorption Spectroscopy using a diode laser was applied for sensitivity enhancement. This method is one of the cavity-based LAS. A diode laser can be used without pulsation. Moreover, it enables us to have enough attenuated power, while providing detection sensitivity comparable to CRDS.

II. Theory

A. Laser Absorption Spectroscopy

The intensity variation dI of a laser beam with intensity I propagating through an absorbing sample, whose absorption coefficient is $k_{\mathbf{v}}$ and length is d_0 , is expressed as follows,

$$dI = -k_{\nu}I_{0}dx \tag{1}$$

The absorption coefficient k_v is the function of the laser frequency and expressed in following Voigt profile.

$$k_{\nu} = K \frac{2}{\Delta \nu_{\rm D}} \sqrt{\frac{\ln 2}{\pi}} \int_{-\infty}^{\infty} \frac{\exp(-t^2)}{y^2 + (x - t)^2} dt$$

$$x = \frac{\sqrt{\ln 2} (v - v_0)}{\Delta v_{\rm D}}$$

$$y = \frac{\sqrt{\ln 2} \Delta v_{\rm L}}{\Delta v_{\rm L}}$$
(2)

Number density of absorption state n_1 is related to integrated absorption coefficient K as,

$$K \equiv \int_{-\infty}^{\infty} k_{\nu} d\nu \approx \frac{\lambda^2}{8\pi} \frac{g_j}{g_i} A_{ji} n_i \cdot$$
(3)

And the translational temperature is related to the Doppler width as follows.

$$T_{tr} = \frac{c^2 \cdot (\Delta v_{\rm D})^2 M_{\rm A}}{8 \ln 2 \cdot k v_{\rm O}^2}$$
(4)

B. Cavity Enhanced Absorption Spectroscopy

In CEAS, only a laser beam whose frequency meets the resonance condition can transmit the cavity. Then, the absorption signal is obtained through integration of the total signal transmitted through an optical cavity as shown in Fig.1. The integrated transmitted signal as the summation as follows,

$$I = I_0 T^2 \sum R^{2n} \exp\{-(2n+1)k_{\nu}d_0\}$$
(5)

where n is the number of reflections in the cavity. The summation can be approximated by an integral over n (for large n) resulting in as follows

$$I = I_0 T^2 \exp(-k_{\nu} d_0) \int_0^\infty \{ R \exp(-k_{\nu} d_0) \}^{2n} dn \,.$$
(6)

Solving this expression, following equation is obtained.

$$\frac{I}{I_0} \approx (1-R)^2 \frac{\exp(-k_v d_0)}{-2\ln\{\operatorname{Rexp}(-k_v d_0)\}}$$
(7)

2 American Institute of Aeronautics and Astronautics Here the mirror loss is neglected. When an absorber is placed in the cavity, the transmitted signal for frequency scan can be obtained as shown in Fig.2.



Fig.1 Predicted transmitted signal for relative frequency

The relationship between the LAS fractional absorption and CEAS fractional absorption for a variation of the mirror reflectivity is shown in Fig.3. For example, when mirrors with 99.9% reflectivity are used for CEAS, fractional absorption is enlarged by three orders of magnitude. The measurement limit is also shown in Fig.3. As shown in this figure, the measurement limit of LAS is about 10^{-3} . So, CEAS is effective in the lower fractional absorption range. In this range, following approximation is valid.

$$1 - \exp\left(-k_{\nu}d_{0}\right) \approx k_{\nu}d_{0} \tag{8}$$

So, effective absorption length is thought as follows.

$$d_{eff} = \frac{d_0}{1 - R} \tag{9}$$



Fig.2 Predicted transmitted signal when the absorption is placed in the cavity.



LAS fractional absorption

Fig.3 Theoretical Enhancement of fractional absorption and measurement limit

III. Experimental setup

The schematic of measurement system is shown in Fig.4. A tunable diode-laser with an external cavity was employed (EOSI DMD845) which is not accompanied with mode-hops in wide modulation width. The laser was scanned over about 20GHz interval at a repetition frequency of 0.35Hz with a function generator (SONY-TEKITRONIX, AGF310) High reflection concave mirrors whose curvature radii are 1m were used for constructing a cavity. A faraday optical isolator (ISOWAVE, I-80-U4) was inserted close behind the laser to reduce the optical feedback. А Fabry-Perot etalon (NEOARK) was used whose free spectra range is 0.75GHz to measure the frequency change.

In CEAS, when the loss in the cavity is larger than absorption, the absorption can't be detected. Therefore, reduction of the loss in the cavity is



Fig.4 The schematic of measurement system for cavity enhanced absorption spectroscopy

important in CEAS. In this paper, a plasma torch is used for testing CEAS method. Because it can generate a plasma flow in the atmosphere, there is no loss in the cavity. Moreover, alignment of mirrors can be always corrected easily. The schematic of the plasma torch and its photo in operation are shown in Fig.5. Its exit diameter, input power and discharge voltage were 1mm, 500W and 20V, respectively. Argon whose flow rate was 1.3slm was supplied from the base of cathode rod. In this paper, a metastable ArI 842.46nm is used as the absorption wavelength line. An argon glow discharge tube (CI TECHNO CO., Ltd) was used for monitoring absorption wavelength of the target line. Its input power, discharge voltage and ambient pressure were 2.0W, 400V and 80Pa, respectively. A bandpass filter (Thorlabs INC, FB840-10) was used to reduce the plasma emission. Its center wavelength is 840nm and full width at half maximum is 10nm. The transmitted laser was detected by a photo detector (Thorlabs INC, DET110).





Fig.5 Configuration of plasma torch (left) and its photo in operation

IV. Results and discussion

A. Cavity mode

There are two cavity modes; on-axis mode and off-axis mode. Fig.6 shows images of transmitted beam observed by CCD camera. From these images, the axis of the laser for each mode was adjusted. Figure 7 shows the resonance signals of on and off axis modes by photo detector looking through the mirrors of the cavity. In on-axis mode, laser beam is reflected on the center of mirrors. However, the resonance is sensitive for the mechanical vibration. So, the transmitted laser intensity is not constant. While in off-axis mode, laser is reflected not only on the center of the mirrors but also on the other several points of mirrors. In other words, rather than a single possible alignment geometry (i.e., the laser on axis with the cavity), a virtually infinite number of stable paths through the cavity can be used. So off-axis mode can be a robust alignment for the mechanical vibration. As a result, the transmitted laser intensity in off-axis mode is more stable than the one in on-axis mode as shown in Fig.7. Stable transmitted laser intensity enables to obtain a profile with low noise. From this reason, the experiments were performed in an off-axis mode.



Fig.6 Images obtained by a CCD looking through the rear mirror of the cavity showing the on-axis cavity alignment patterns (top) and off-axis one (bottom).

Fig.7 On-axis mode and off-axis mode resonance signal by photo detector looking through the rear mirror of the cavity.

B. The measurement results of single-pass Laser Absorption Spectroscopy

First, the plasma torch was diagnosed by single-pass LAS. LAS signals in plasma torch flow and glow discharge tube plasma and etalon signal are shown in Fig.8. The absorption profiles were fitted by Voigt profiles as shown in Fig.9. From this figure, the profile of glow discharge tube was almost expressed as gauss profile. While, the profile of plasma torch had the pressure broadening and shift because of the operation under atmospheric pressure. From these Doppler width and integrated absorption coefficient, the translational temperature and the number density were deduced as 640K, $1.1 \times 10^{17} \text{m}^{-3}$ for the glow discharge tube and 12,000K, $5.3 \times 10^{17} \text{m}^{-3}$ for plasma torch, respectively.



Fig.8 LAS signal of plasma torch and glow discharge tube and etalon signal

Fig.9 Absorbance of plasma torch and glow discharge tube with Voigt fitting results.

And then the absorption profiles of the plasma torch at several positions from the exit were measured as shown in Fig.10. From this figure, the translational temperature and meta-stable ArI number density can be obtained as shown in Table.1.



Fig.10 LAS signal of plasma torch at several positions from the exit

Distance from the exit, mm	Translational temperature ,K	Meta-stable ArI number density, m ⁻³
0	12,000	$5.3 \ge 10^{17}$
0.5	8,500	$2.8 \ge 10^{17}$
1	5,700	1.6 x 10 ¹⁷

Table.1 Measured translational temperature and meta-stable ArI number density of plasma torch at several positions from the exit

C. The measurement result of Cavity Enhanced Absorption Spectroscopy

A CEAS signal of plasma torch at 1mm from the exit, a LAS signal of glow discharge tube and an etalon signal are shown in Fig.11. The averaged CEAS profile for 85s (30 sweeps) is shown in Fig.12. In Fig.12, a line shows the absorption profile with assumed effective absorption length ratio d/d_0 to the one of single-pass LAS d_0 . As a result, the effective absorption length ratio of this cavity is estimated about 400.

In this cavity, the reflectivity of the mirrors is more than 99.95%. So, the theoretical effective absorption length ratio of this cavity is estimated to be 2000. This reason for the difference between the theoretical value and the practical one is caused by off axis mode. Then the laser in the cavity is distributed about $2.5 \times 10^{-3} \text{m}^{-2}$, while the absorber is distributed about $1 \times 10^{-4} \text{m}^{-2}$. As a result, more than 90% laser is passed without absorption. Because of these reasons, the practical effective absorption length is smaller than the ideal one. However, in our research sensitivity enhancement more than two orders of magnitude is needed. And when effective absorption length ratio is large, the transmitted laser intensity is almost zero. Then whole absorption profile is not obtained. So this practical value is convenient.



Fig.11 CEAS signal of plasma torch, LAS signal of glow discharge tube and etalon signal

Fig.12 Averaged CEAS profiles of plasma torch at 1mm from the exit and estimated profiles with assumed effective absorption length as 400

In Fig. 13, the normalized CEAS profile of plasma torch at 2mm from the exit averaged for about 85s (30 sweeps), where the absorption signal could not be obtained by LAS, is shown. From this figure, the absorbance at 2mm can be obtained as shown in Fig.14. And the translational temperature and meta-stable ArI number density are obtained as 4,500K and 8×10^{15} m⁻³, respectively.



0.8 0.7 0.6 0.5 0.5 0.4 0.3 0.2 0.1 0 -15 -5 5 15 Relative frequency, GHz

Fig.13 Averaged CEAS profiles of plasma torch at 2mm from the exit

Fig.14 Averaged CEAS absorbance of plasma torch at 2mm from the exit with Voigt fitting result

V. Conclusion

- 1) The system for the Cavity Enhanced Absorption Spectroscopy was developed by added high reflection mirrors to the system for the conventional single-pass Laser Absorption Spectroscopy and applied to a plasma torch.
- 2) Compared with the conventional system of the single-pass LAS, the sensitivity of this CEAS system was successfully increased by two orders of magnitude.
- 3) This ratio of sensitivity enhancement is convenient for applying to plasma wind tunnels. Because when a cavity with larger effective absorption length ratio is used, whole absorption profile could not be obtained.

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VII. References

¹Auweter-Kurtz, M., Kurtz, H. and Laure, S. "Plasma Generators for Re-Entry Simulation", *Journal of Propulsion and Power*, 1996, Vol. 12, No. 6, pp.1053-1061.

²Matsui, M., Takayanagi, H., Oda, Y., Komurasaki, K., Arakawa, Y., "Performance of arcjet-type atomic-oxygen generator by laser absorption spectroscopy and CFD analysis", Vacuum, 2004, Vol.73, 3-4, pp.341-346

³Matsui, M., Komurasaki, K., and Arakawa, Y., "Laser Diagnostics of Atomic Oxygen in Arc-Heater Plumes", 40th AIAA Aerospace Science Meeting and Exhibit, 2002, AIAA 02-0793

⁴H. Takayanagi, Matsui, M., Komurasaki, K., and Arakawa, Y., "Sensitivity Enhancement in Laser Absorption Spectroscopy for the Diagnostics of High Enthalpy Flows," 43rd AIAA Aerospace Science Meeting and Exhibit, 2005, AIAA-2005-0831

⁵S. Kim, J. B. Jeffries and R. K. Hanson, "Measurements of Gas Temperature in the Arc-heater of a Large Scale Arcjet Facility using Tunable Diode Laser Absorption", 43rd AIAA Aerospace Science Meeting and Exhibit, 2005, AIAA-2005-0900

⁶O'Keefe, A., Deacon, D. A. G., "Cavity Ring-down optical spectrometer for absorption measurements using pulsed laser sources", *Review of Scientific Instrument*, 1988, Vol. 59, No.12, pp.2544-2551.

⁷A.J. Romponi, F.P. Milanovich, T. Kan and D. Deacon, "High sensitivity atmospheric transmission measurements using a cavity ringdown technique", *Applied Optics*, 1988, Vol. 27, No.22, pp.4606-4608

⁸R.T. Jongma, M.G.H. Boogaarts, I. Holleman and Geradr Meijer, "Trace gas detection with cavity ring down spectroscopy", *Review of Scientific Instruments*, 1995, Vol.66, No.4, pp.2821-2828

⁹D. Romanini, A.A. Kachanov, N. Sadeghi and F. Stoeckel, "CW cavity ring down spectroscopy," *Chemical Physics Letters*, 1997, Vol. 264, pp.316-322

¹⁰J.B. Paul, L. Lapson and J.G. Anderson, "Ultrasensitive absorption spectroscopy with a high-sinesse optical cavity and offaxis alignment", *Applied Optics*, 2001, Vol.40, No.27, pp.4904-4910