Application Examples of a Rapid and Simplified Coherent Anti-Stokes Raman Scattering (CARS) Technique

S.Fujii
Department of Aeronautics
University of Osaka Prefecture
M.Gomi
Thermofluid Dynamics Division
National Aerospace Laboratory
7-44-1 Jindaiji-Higashi
Chofu, Tokyo 182
Japan

ABSTRACT

A new spectroscopic concept for the rapid determination of temperature and oxygen concentration by CARS(Coherent Anti-Stokes Raman Scattering) was described. The ratio of two spectral regions in the broadband Q-branch spectrum was detected by photomultipliers in a monochromator, which ratio depends on temperature and species concentration. The comparison of the measured data with theory was made using a flat flame burner and an electric furnace, with reasonable results. A new beam splitting method was introduced. The combination of the spectroscopic concept and the optical techniques will make the CARS measurement system rapid in data processing and simple in optical parts. Application examples are shown.

INTRODUCTION

CARS(Coherent Anti-Stokes Raman Scattering) techniques have been widely applied in combustion diagnostics. For CARS thermometry, nitrogen molecules are often chosen because of their large concentrations available as an inert gas. In combustion gases, the concentration of diatomic molecules such as O_2 and CO may be easily determined by CARS because of their simple theoretical spectrum. Most measurements have been carried out by multiplex CARS where the broadband of ω_2 is employed.CARS has many advantages in its application. It has excellent discrimination against luminous or fluorescence backgrounds. In contrast, the present authors have found a few drawbacks during a series of practical applications. By use of one optical multichannel analyzer, it takes appreciable time to record the intensity in multichannels and then compare the measured data with theory. As the repetition rate of the laser increases, it will be unable to furnish the instantaneous time-varying temperature and concentration on a real-time basis. The acquisition and storage of broadband spectral profiles may need a photodetector and analyzer with many channels and a monochromator with significantly high resolution. Such devices are quite expensive. This paper proposes a new method to simplify the spectroscopic procedures, which results in real-time determinations of temperature and concentration. Various optical techniques are also introduced. The combinations of the new spectroscopic concept and these new optical techniques will make the CARS system fast in data processing and simple in optics, with a remarkable decrease in optical and electronic instruments costs. Application examples are shown.

RAPID TEMPERATURE DETERMINATION BY NITROGEN CARS

The CARS intensity depends on the squared third-order susceptibility of molecules in the gas, which can be written as

$$|\chi^{(3)}|^2 = |\chi' + i\chi'' + \chi_{NR}|^2 \qquad (1)$$

Nitrogen molecules are often chosen for CARS temperature measurements because of their large concentrations in hot and chemically reacting flows. For such large populations, the resonant symmetric term $|\chi''|^2$ is dominant and other terms $|\chi'|^2$, χ_{NR}^2 and $2|\chi'|\chi_{NR}$ are negligible. The spectral profiles of $|\chi''|^2$ depends only on temperature through the Maxwell-Boltzman equilibrium distribution for the Q-branch spectrum. The comparison of experimental profiles with theory gives information on temperature. However, it takes some time to sweep the voltages of intensity in the multichannels and then compare the measured data with theory in the whole Q-branch range. Because of the amount of time necessary to do this comparison, real-time measurements are not possible. Instead of using the whole Q-branch spectrum, the ratio of partial integrals such as hot/cold bands, warm/cold bands and their combinations have been reported (1). However, the integrations were performed numerically on the measured data after

the optical multichannel processes, which still did not give the real-time data reduction. The peak ratio of hot/cold bands might provide one possibility for speeding up the process but with rather large errors(2). As another alternative, a new spectroscopic concept is proposed herein.

The basic idea starts by splitting the CARS signal using spectrometer exit slits #1 and #2, as shown in figure 1. By changing the two slit widths, various intensity ratios of slit #1 to #2 could be optically obtained.

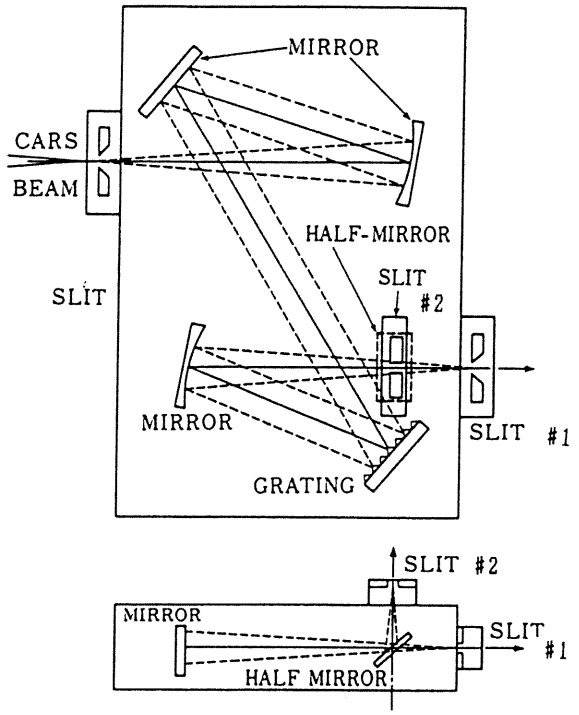


Fig. 1 Monochromator with half mirror inserted

A propane-air mixed flat flame burner was employed in the experiment. By use of the burner, a nitrogen spectrum at high temperature was obtained by scanning the monochromator. The results are shown in figure 2. We made an average of over 100 pulses. The best fit curve was obtained by adjusting the value of the slit function in the computer code, as indicated in figure 2. The value obtained from the best fit was 5.5 cm^{-1}. The spectra at three temperatures with this slit function were convoluted in figure 3, where the widths of slits #1 and #2 are also illustrated along the horizontal axis. The intensity ratio for slit #2 to slit #1 was calculated in the temperature range from 300K to 2500K and plotted in figure 4 and is denoted by CASE(1). The width of slit #1 was selected to give the curve sensitivity to the temperature change. To see the effect of slit width, the width of slit #1 was slightly widend. The result is drawn in figure 4 as CASE(2).

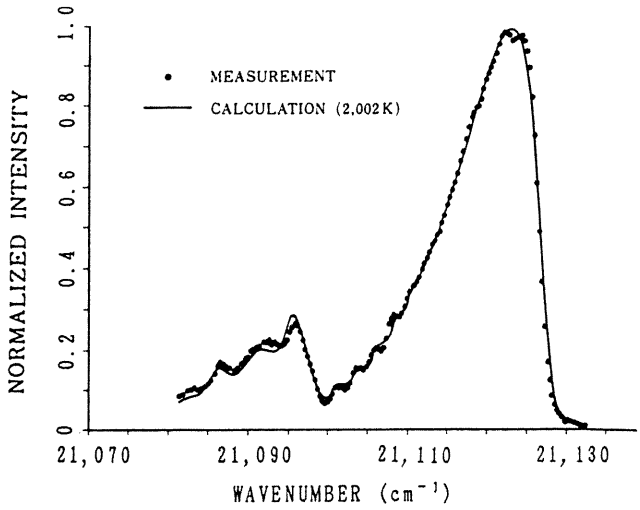


Fig. 2 Nitrogen Q-branch spectrum at high temperature

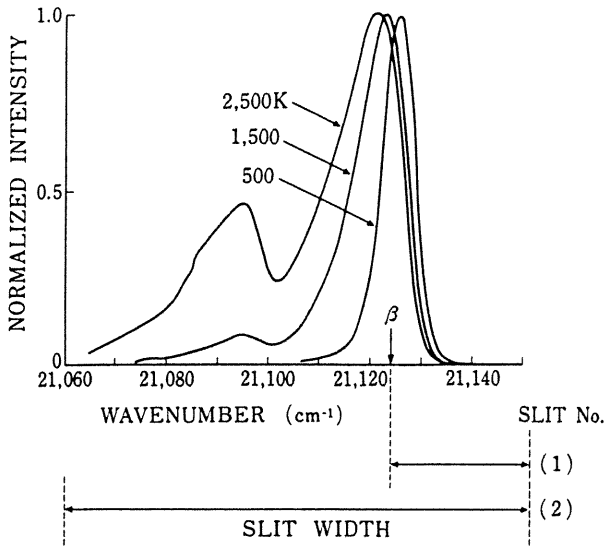


Fig. 3 Calculated spectrum and relationship between slit width #1 and #2; #1=21122.2～21155 cm^{-1}

To compare the present method with the conventional whole-spectrum method, simultaneous measurements were performed. The test setup shown in figure 5 consisted of two optical paths. The 700 mJ/pulse Nd:YAG laser with 20Hz repetition rate, homemade dye oscillator, flat flame burner and computer were common to both paths. The CARS signal was generated within the burner and split by a half mirror. One of the CARS signals was passed through the monochromator with a half mirror inserted, detected by a photomultiplier followed by a sample-hold circuit and then fed to the computer. This is the path for the new concept. The other signal was processed by the monochromator, silicon-intensified array detector and optical multichannel analyzer. The latter describes the lineup for a standard CARS measurement. It is

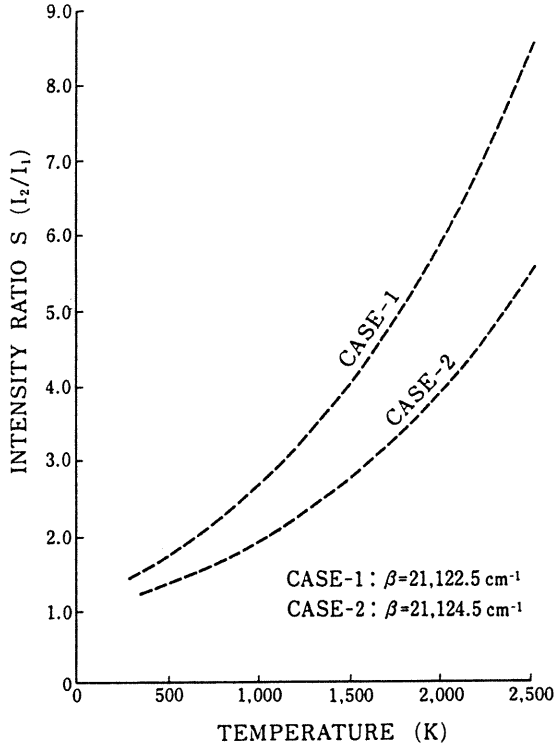


Fig. 4 Calculated intensity ratio; for CASE(2), slit #1 was increased by 2 cm^{-1}

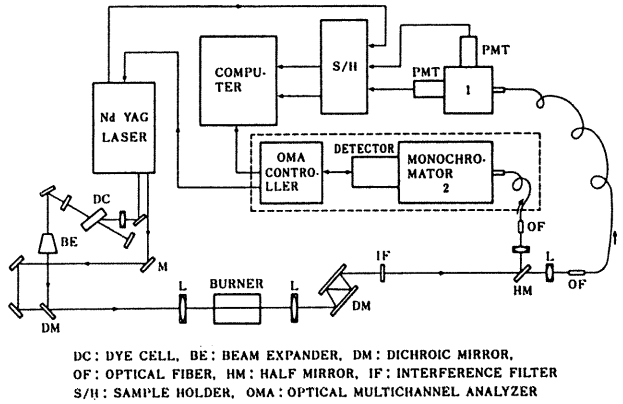


Fig. 5 Experimental setup for parallel data acquisition by conventional and proposed method

further noted that the monochromator associated with the two photomultipliers had much less resolution (0.03 nm,FWHM), compared to that (0.007nm) of the monochromator used in the conventional arrangement.

Extensive and rigorous comparisons were made for the measured temperatures by the conventional CARS, and by the two-photomultiplier method. The results are summarized in figure 6. The straight line relationship validates the new method. It was not sensitive to the change of slit widths within the data range of CASE(1) and CASE(2). Note that all data were obtained from the average over 100 pulses.

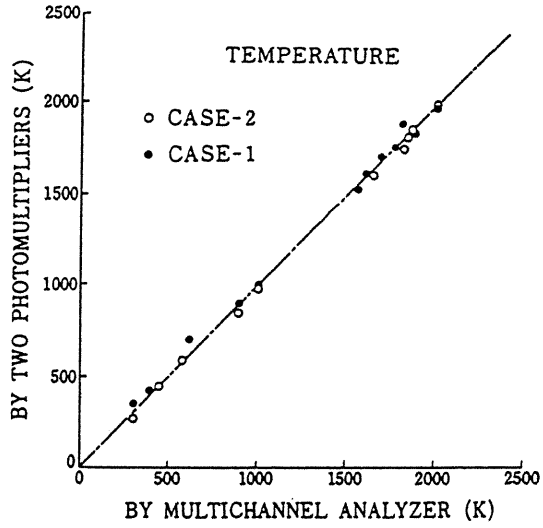


Fig. 6 Comparison of two temperature measurements

RAPID CONCENTRATION DETERMINATION BY OXYGEN CARS

In a combustion gas, oxygen is sometimes detected in low concentrations because it is reactive. In such a situation, the two terms of $|\chi'|^2$ and $|\chi''|^2$ are neglected and the susceptibility can be written as,

$$|\chi^{(3)}|^2 \sim \chi_{NR}^2 + 2|\chi'|\chi_{NR} \qquad (2)$$

Equation (2) indicates a significant domination of the nonresonant term χ_{NR} which implies a difficulty in detecting low concentrations. One method of overcoming this difficulty is to use a polarization technique for rejecting χ_{NR}. However, the disadvantages in using polarized CARS are twofold. One is associated with the considerable attenuation of signals under the conditions of extremely low signal-to-noise ratio. The other is that the measurement system is further complicated by the addition of expensive polarization devices. Instead of rejecting the nonresonant term, the intensity ratio method was again employed. Figure 7 shows a typical computed O_2 spectrum of the Q-branch at a concentration of 3% in volume for a temperature of 1500K. The same slit function of 5.5 cm^{-1} as that obtained in the temperature experiment was used with a nonresonant susceptibility of $10.97\times10^{-18}\times(273.15/T)cm^3/erg$. The susceptibility given by equation (2) depends on both temperature and concentration. We first calculated the ratio of numericl integrals #2/#1 and next those #3/#1 at various temperatures and concentrations. The results are plotted in figure 8 and 9. The integral over #1 represents the magnitude of χ_{NR}. Therefore, the integral ratio shown in figure 8 can be considered the hot/nonresonant ratio, which is rather insensitive to the change

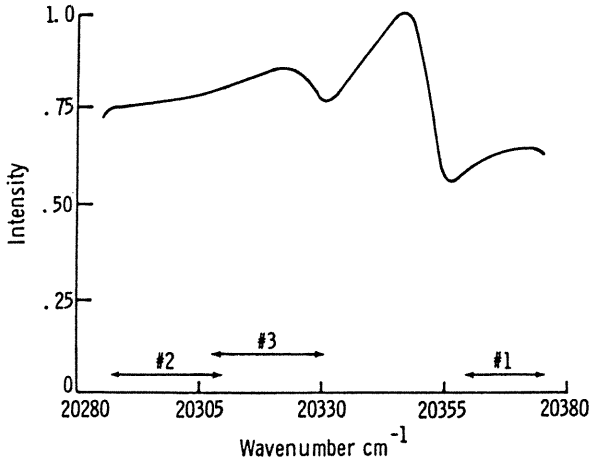


Fig. 7 Oxygen Q-branch spectrum at 3% concentration in volume for 1500K:97% nitrogen

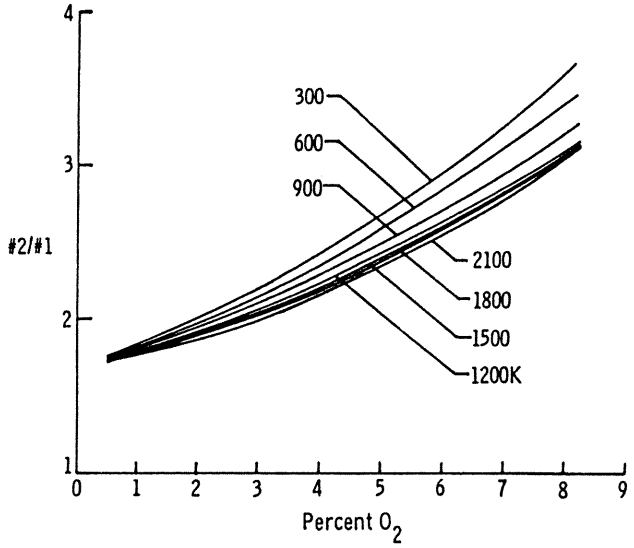


Fig. 8 The calculated intensity ratio of slit #2 to #1 ; #1=20360～20375 cm^{-1}, #2=20287～20312 cm^{-1}

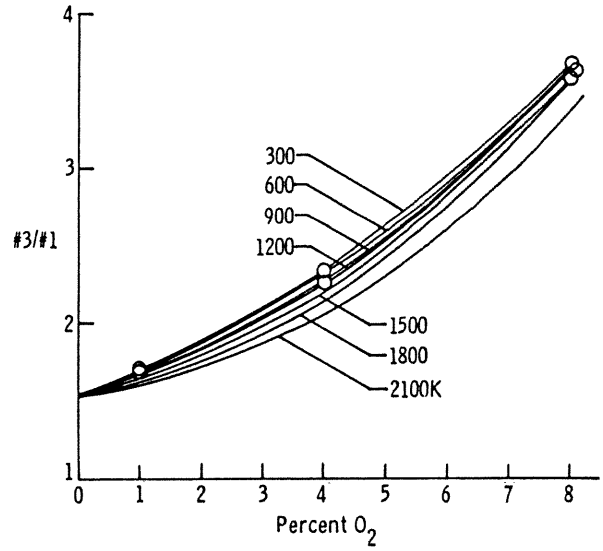


Fig. 9 The calculated and measured intensity ratio of slit #3 to #1 ; #3=20307～20330 cm^{-1}

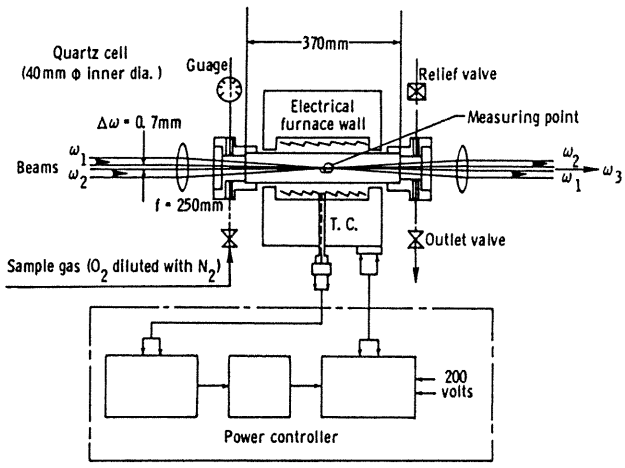


Fig. 10 Electric furnace and measurement setup for O_2 detection

of temperature beyond 1200K but still sensitive to the concentration variation. On the other hand, the ratio of #3 to #1 in figure 9 is the warm/resonant ratio and is useful for concentration determinations below 1200K.

A small electric furnace was used to compare the measured and calculated ratios and is illustrated in figure 10. Using pure nitrogen gas, we first made the CARS temperature measurement within the furnace and determined the relationship between the gas temperature and the wall thermocouple prior to all tests. The uniformity of gas temperature within the furnace space was also ascertained. In the oxygen detection tests, the O_2 gas, diluted by N_2, was introduced into the furnace and heated up to the predetermined temperature. At an O_2 concentration of 8%, the data were obtained in the temperature range of 300K up to 900K at 100K steps. At 4%, three data points at 300K, 600K and 900K were collected. Data of 1% concentration were obtained only near room temperature (300K) and the maximum temperature of 900K. The mreasured data are plotted in figure 9. All data were obtained from the 100 pulse ensemble average. The agreement with theory was reasonable. It was not possible to provide measured data beyond 900K due to the furnace capability.

It is further noted that the present method of determining temperature and concentration assumes an equilibrium state between the rotational and vibrational degrees of freedom in the gas to be measured. The application to turbulent flames would be the next step including a rigorous evaluation of measurement uncertainty.

NEW BEAM SPLITTING METHOD

In the CARS measurement, a split of the ω_1 beam is sometimes required. It is proposed that a

prism be used to separate the beam into two hemicircles, as illustrated in figure 11. It is of interest to note that the laser beam ω_2 appeared as a hemicircle as it was generated in the dye cell with the hemicircle pump beam. This fact is advantageous for the new alignment using the hemicircle of ω_1 and ω_2. One more feature is that the transverse movement of the prism will change the beam split ratio. In other words, the intensity ratio of ω_2 to ω_1 can be varied arbitrarily to obtain the optimum signal, which would be particularly advantageous under adverse environments.

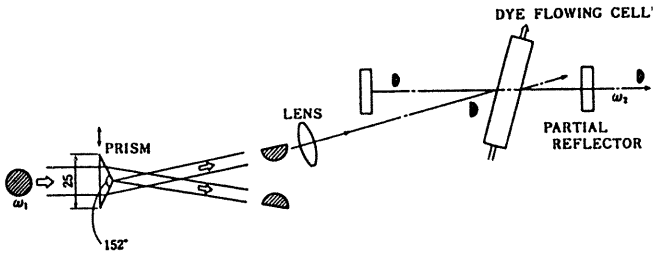


Fig. 11 Beam split by prism

APPLICATION EXAMPLES

One of the practical applications was made for the determination of temperature charactristics in a pulse burner developed for home appliance, in figure 12. Figures 13(a) and 13(b) shows the probability density functions of the measured data in axial and normal directions, respectively.

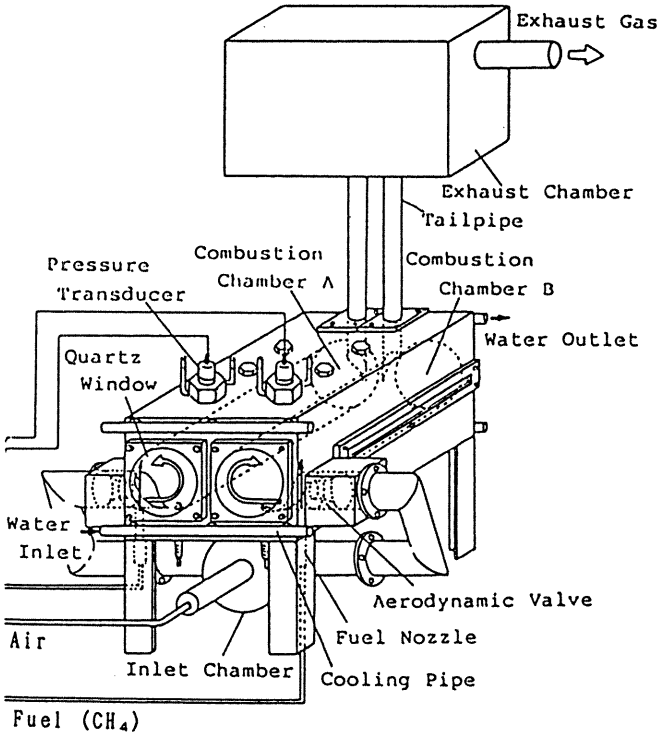
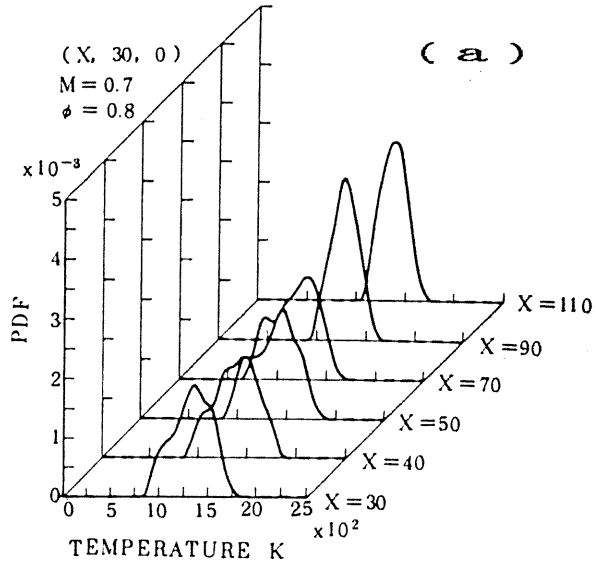


Fig. 12 Schematic of twin valveless pulse combustor



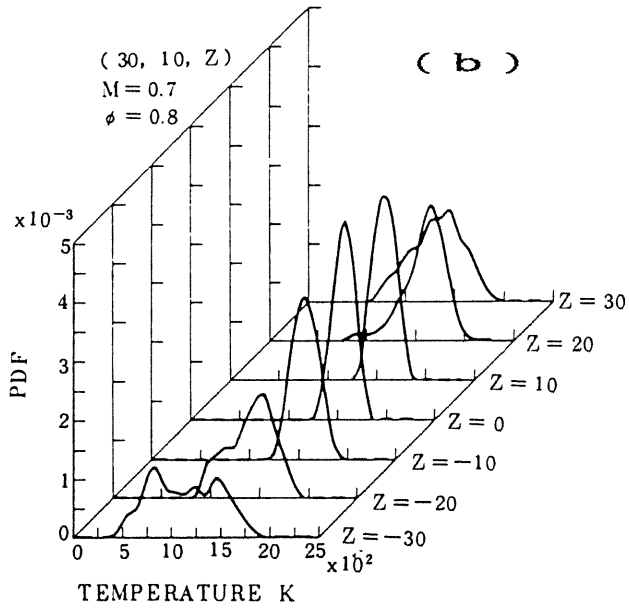


Fig. 13 Probability density functions of temperature
(a) Axial direction
(b) Normal direction

Another application example is shown in figure 14, where the hot airflow was issued into the cold main stream with a Mach number of 2. Figure 15 illustrates the temperature histograms as a result of hot/cold airflow mixing at high speeds. Using the flat flame burner, figure 16, we measured the oxygen concentrations at the various heights, figure 17. In this case, the temperature information was furnished by a thermocouple prior to the tests.

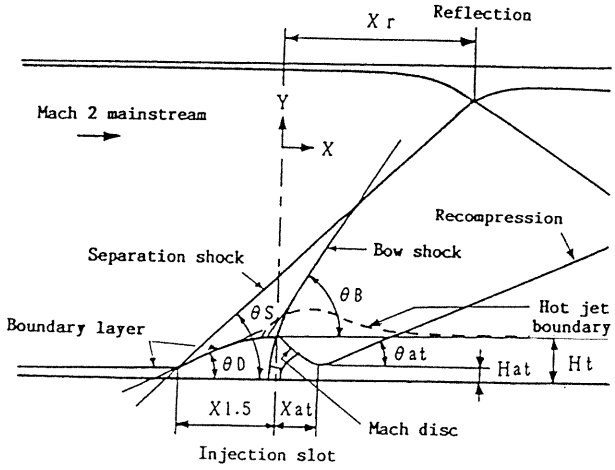


Fig. 14 Schematic-flowfiled of supersonic flow mixing (2-D normal injection)

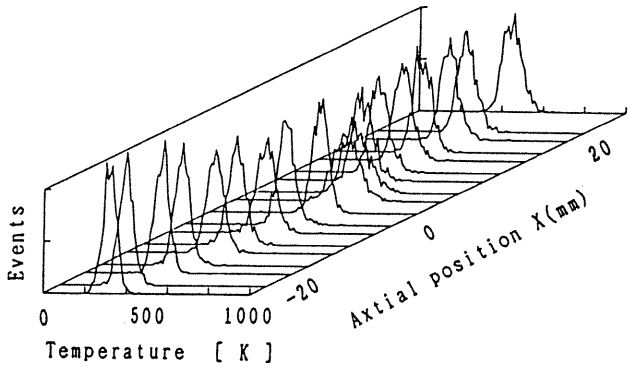


Fig. 15(a) Temperature histograms (near wall)

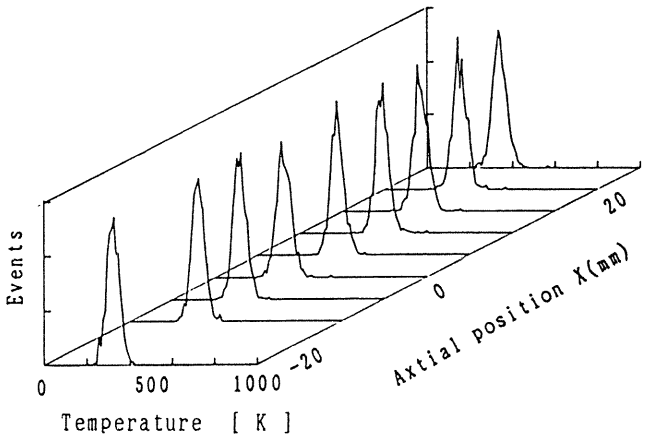


Fig. 15(b) Temperature histograms (some distance from wall)

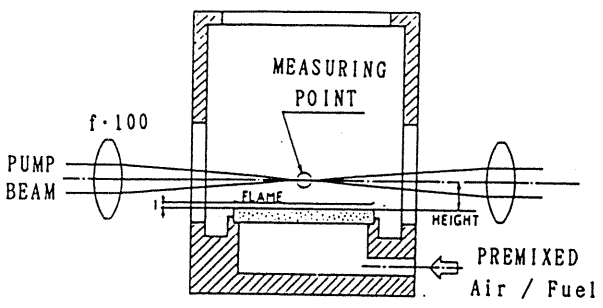


Fig. 16 Flat flame burner

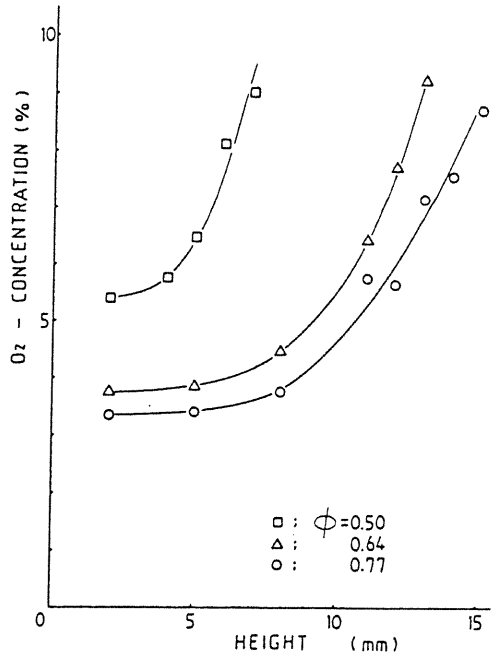


Fig. 17 Measured oxygen concentration above the burner surface

NOMENCLATURE

T	:gas temperature, in kelvin
$\chi^{(3)}$	:third-order susceptibility
χ'	:unsymmetrical term
χ''	:symmetrical term
χ_{NR}	:nonresonant term
ω_1	:pump frequency
ω_2	:Stokes frequency
ω_3	:CARS frequency
ϕ	:stoichiometric ratio

$i^2=-1$

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