

Two-Dimensional Spectroscopic Analysis of a Flame Using Highly Preheated Combustion Air

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The effect of highly preheated combustion air on the combustion characteristics of propane flames is presented. Data have been obtained on the spatial distribution of emission intensity from selected radicals, fluctuations of emission intensity, and flame temperature using several diagnostic techniques. Several propane and high-temperature air flames, produced with the crossflow diffusion of gas into highly preheated combustion air (obtained by a regenerative combustion facility) having normal oxygen content and reduced oxygen content (<21% by volume) have been examined. The diagnostics used include a UV-charge coupled device (CCD) camera integrated with an interference filter, a high-speed video camera, array of thermocouples, and gas analyzers for NO_x and O₂. The UV-CCD camera fitted with the desired optical interference filter provided the spontaneous emission signature of the flames produced. Fast Fourier transform applied to the video camera recorded image provided information on the two-dimensional distribution of emission intensity fluctuation. The emission results show that the intensity level of OH and C₂ radicals in the flame with highly preheated and lower oxygen concentration air is much lower than the corresponding normal air flames. The fluctuations of emission intensity with highly preheated combustion air are significantly lower in the high-temperature combustion region of the flame. The temperature measured from the ratio of the emission intensities had much higher spatial resolution than those obtained with the conventional thermocouple array. The results indicate that preheated air drastically reduces both the temperature gradient and temperature fluctuations in the flames. NO_x measurements were made in the exhaust gas using a chemiluminescence analyzer. The results showed that reducing the oxygen concentration in the high temperature combustion air reduces NO_x emission levels in the exhaust gas.

Nomenclature

A	= Einstein's transition probability for spontaneous emission, s ⁻¹
E	= excitation level energy, J
g	= statistical weight
h	= Planck's constant, 6.626176 × 10 ⁻³⁴ J/Hz
I	= spectral intensity of spontaneous emission, W/sr
$I_{av}(x,y)$	= average spectral intensity, W/sr
K	= $vgA/Q(T)$, s ⁻²
k	= Boltzmann's constant, 1.380662 × 10 ⁻²³ J/K
N	= number density, combustion radicals/m ³
$P_{f(x,y)}$	= intensity at frequency f in the frequency-domain spectrum after FFT
$Q(T)$	= partition function
$S_f(x,y,sp)$	= spectrally observed flame fluctuation, W/sr
T	= vibrational temperature, K
ν	= frequency, Hz
ϕ	= equivalence ratio

Introduction

THE use of highly preheated air as an oxidizer in regenerative combustion systems is of significant interest in all types of industrial furnaces, boilers, and power systems.^{1,2} This technology has been demonstrated to provide significant energy savings and reduced levels of pollutants emission as compared to other available advanced technologies for fossil and derived fuels. Furthermore, the results have shown that the size of the equipment is smaller for the same throughput, or higher throughput of materials can be obtained for same size of the equipment.^{1,2} Energy savings directly translate to reduction of CO₂ emission to the environment. The lower flammability limit of the mixture is significantly extended to the fuel-lean combustion zone.¹⁻⁵ At very low oxygen concentration, flameless oxidation of certain fuels has been shown.^{3,5} Furthermore, the mixture can be combusted with air containing very small concentration of oxygen (only about 1–2% or less). Fuels of very low heating value and very fuel-lean mixtures can also be burned under high-temperature air combustion conditions.⁵ This reveals that gas streams containing only a small amount of fuel can be burned rather than using some gas cleanup device for pollution control of the gas stream. This technology also allows one to use flue gases containing a very small amount of oxygen concentration as the oxidizing gas. A direct benefit of this technology is in the high-enthalpy recovery from the exhaust gases of any furnace or power plant, which also reduces the requirements on the amount of excess air and input fuel. In addition, reduced oxygen concentration in air has the capability to reduce NO_x emission from the combustion of hydrocarbon fuels.

Several fundamental studies on highly preheated air combustion have shown that the features of such flames are significantly different than those obtained with normal air at room temperature or some moderate degree of air preheat.¹⁻⁵ The global and detailed characteristics of the flames are significantly different with high-temperature combustion air. Under certain conditions, flameless or colorless oxidation of the fuel has been observed.⁵ The flame color

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has been observed to be green, purple, or orange under certain operational conditions.^{3,5} The uniformity of temperature within the combustion zone is high while the temperature fluctuations are extremely low in the entire burning zone. The thermal field uniformity and flame stability with high-temperature air combustion are discussed in Ref. 6.

In a previous study by the authors,⁷ spontaneous emission analysis was developed and applied for nonintrusive observation of a methane-air diffusion flame using highly preheated combustion air in a small burner. It was shown that flame uniformity and stability are significantly enhanced with highly preheated combustion air.^{6,7} These important characteristic features of flames with high-temperature combustion air, obtained using regenerative combustion systems, are finding many new applications in almost all kinds of furnaces and boilers, fuel energy conversion and utilization systems, and power and propulsion systems.⁸ In this study, two-dimensional distributions of temperature and flame fluctuation have been obtained for liquified petroleum fuel (LPG), which is mostly propane, flames formed with the crossflow diffusion of propane into highly preheated combustion air in a regenerative combustion furnace. The flame features have been examined using several diagnostic techniques, such as an UV-charge-coupled device (CCD) camera coupled with an interference filter, a high-speed video camera, an array of thermocouples, and an NO_x analyzer. The results obtained are discussed along with the thermal field uniformity and fluctuations of the flame.

Experimental Apparatus

A schematic diagram of the experimental apparatus used is shown in Fig. 1. The regenerative test facility, designed and fabricated by Nippon Furnace Kogyo (NFK) Company, Ltd., Japan, provided the high-temperature combustion air. The facility consists of two combustion chambers, each fitted with ceramic (Cordierite) honeycomb regenerator [Fig. 1(8)] and controllers for flow and switching sequence [Fig. 1(10)]. The temperature of preheated air is measured by a thermocouple (R type) located above the regenerator [Fig. 1(6)]. The temperature of the preheated air in the test section reduces temporally after the flame ignition at a rate of 4 K/s¹. The high-temperature combustion air can also be produced on a continuous basis using modified version of this facility, also designed and fabricated by NFK. The continuous-type high-temperature air facility is now available at the University of Maryland Combustion Laboratory. Further details on the furnace and combustion system are given in Ref. 2. The fuel gas used for all of the results presented here was liquid petroleum gas (LPG), which consisted of propane (>90%) and butane plus butylenes (<10%). The flow rate for the fuel gas was held constant at 0.05 m³/h at normal temperature and pressure

(NTP) conditions. The fuel gas was injected from a 1-mm-diam. nozzle, located in the firebrick, in a direction normal to the preheated airflow. This air was preheated via its passage through the ceramic honeycomb regenerator. The maximum temperature to which the air could be preheated with this facility was about 1200°C. In this study the oxygen concentration in air was regulated at either 21.0% (which provided the equivalence ratio of $\phi = 0.08$), or 4.0% (which provided the equivalence ratio of $\phi = 0.41$), obtained by diluting the air with nitrogen. Other oxygen concentration in air could easily be obtained with the facility by mixing the appropriate amounts of the two gases. The total flow rate of air plus N_2 was maintained constant at 15 m³/h at NTP. The ratio of air-to- N_2 flow rate was 1 : 4.

The UV-CCD camera [Fig. 1(4)] (SBIG Model ST-6UV) consisted of a UV-CCD detector having 241×250 pixels and a quartz lens (Nikon UV, focal length, $F = 105$ mm). The light signal, after passing through an interference filter [Fig. 1(3)], was detected by the UV-CCD camera. This arrangement provided spontaneous emission from only selected radical species. Four interference filters have been used to detect OH (306 nm), CH (431 nm), C_2 (471 nm), and C_2 (515 nm). The background emission was measured between the Swan bands at 490 nm. In each case, the background emission was subtracted from the detected signal. The temperature of the preheated air reduces temporally at a rate of 4 K/s due to the cooling down of the heat accumulator with the incoming air. To reduce the experimental uncertainty and enhance reproducibility of results caused by this change, the shutter of the CCD camera was opened for 0.5-s time duration after 2 s from the flame ignition.

The high-speed video camera [Fig. 1(5)], having the highest time resolution of 25,000 frames/s, incorporated a CCD detector having 192×239 pixels (Kodak Model UV). The shutter speed of the video camera was set to 5 ms. The framing speed of the camera was 125 frames/s. This high-speed video camera without any optical filter provided the means to detect emission from the flame over a wide range of wavelengths. The total measurement time to record 1091 frames was 8.73 s. Fast Fourier transform (FFT) was then applied to each 128 set of 1091 frames, to give frequency-domain spectra, which then allowed one to construct the two-dimensional distributions of flame fluctuation. The lens [Fig. 1(2)] focuses the flame image from a region of 20×25 cm to 1/10th of the original size. The CCD camera or the high-speed video camera provided information on the focused flame image.

The measured spontaneous emission from C_2 (at 471 and 515 nm), CH (at 431 nm), and OH (at 306 nm) radicals has been measured over very narrow wavelength bands. These chemical species are amongst the important radicals that provide key role during the combustion process. Figure 2 shows a typical spectrum of propane-air flame and the emission peaks from these chemical species. The two C_2 bands

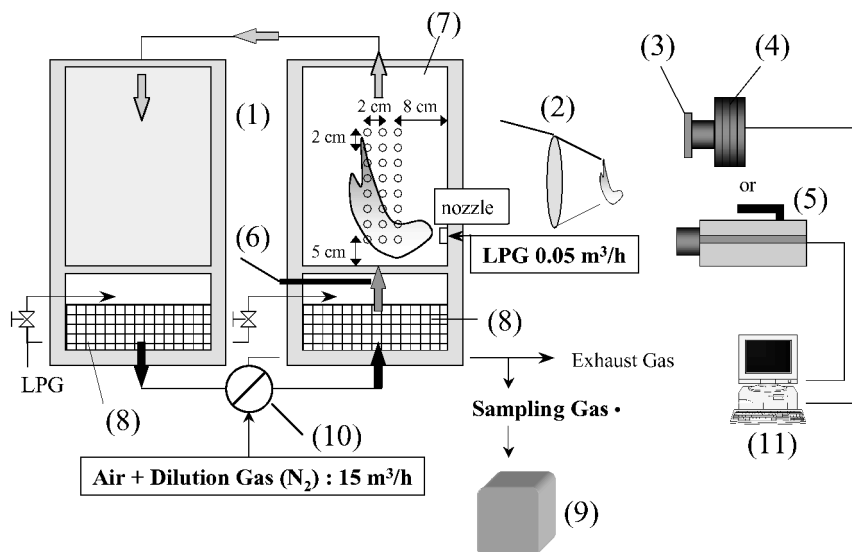


Fig. 1 Schematic of regenerative test furnace facility used for providing the highly preheated combustion air and arrangement for the measurement: 1) test furnace, 2) focusing lens, 3) optical interference filter, 4) UV-CCD camera, 5) high-speed video camera, 6) thermocouple for measurement of preheating temperature, 7) window for observation, 8) regenerator, 9) NO_x analyzer, 10) four-way valve, and 11) microcomputer.

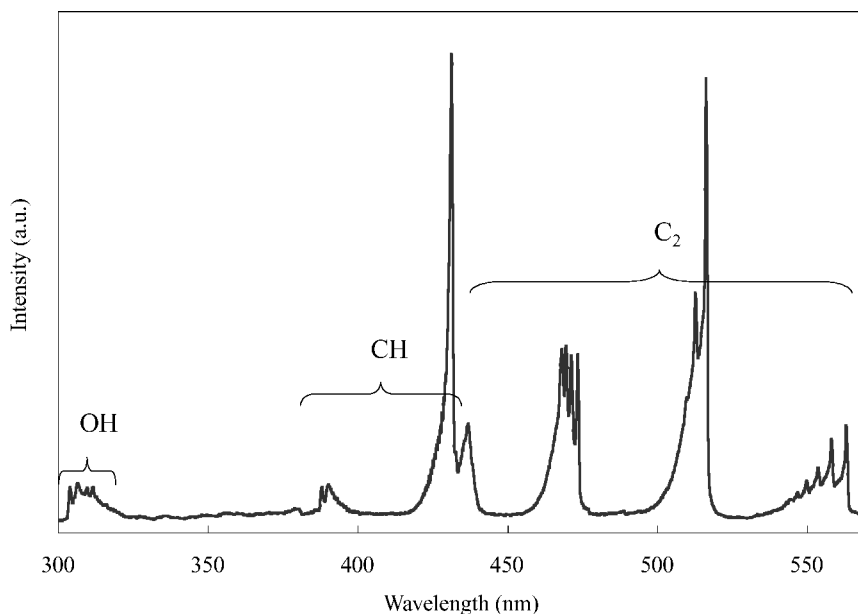


Fig. 2 Typical spectrum of propane-air flame.

(at 471 and 515 nm) were used to calculate the vibrational temperature as well as the two-dimensional distribution of temperatures in the flame, based on a modified two-line method.^{6,7}

In flames with temperatures below 2000 K, the vibrational temperature approximates the gas temperature measured with the thermocouple, or in some cases the overall temperature of the translational, vibrational, and rotational states. This condition is the so-called local thermal equilibrium (LTE). However, there may be non-LTE conditions due to transfer of excess reaction energy to a certain system, for example, the vibrational system. By taking into account such a case, the relationship between the measured temperature with a thermocouple and temperature obtained from the spectral intensity ratio of the two C₂ bands has been obtained. When this calibration curve is used, the band ratio was converted to the thermocouple temperature.

An array of 8 R-type Pt-Pt/Rh thermocouples of 0.1 mm diameter was placed in the combustion chamber. This array of thermocouples was used to measure temperatures at 24 points in the combustion chamber using three consecutive measurements. The calibration and correction of the thermocouples were conventionally made using a radiative heat balance equation between the thermocouples and the combustion chamber wall, respectively.

The NO_x concentration in the exhaust gas was measured with the NO/NO_x chemiluminescence gas analyzer [Fig. 1(9)] having a response time of <30 s (Best Sokki company Model BCL-611A).

Data Processing for Calculating the Flame Fluctuation

A microcomputer [Fig. 1(11)] acquired the signal from the high-speed video camera. The data were then postprocessed using computer programs written in C-language (Turbo-C version 2). The average values were used to construct two-dimensional distributions of the spectral intensity. FFT was applied to the 128 temporally resolved data for the signal intensity at each pixel location of the high-speed video camera. The intensities of the frequency-domain spectra at certain frequency were used to construct the two-dimensional distribution of flame fluctuation. The spectrally observed flame fluctuation $S_{f(x,y),sp}$, at a certain frequency f and position (x, y) was then calculated from

$$S_{f(x,y),sp} = P_{f(x,y)} / I_{av(x,y)} \quad (1)$$

where $P_{f(x,y)}$ is the intensity at frequency f in the frequency-domain spectra that is obtained after the FFT processing of the time series of pixel data (called the time-domain data). The I_{av} is the average spectral intensity. The same algorithm was applied to the time-domain data of the thermocouples to yield temperature fluctuation and its distribution in two dimensions.

In a previous study, the spectroscopic temperature was determined on the basis of a two-line method of the C₂ emission band (Swan band).⁶ However, most of thermal calculations have been based on temperatures measured using thermocouples. Therefore, it is convenient to convert the spectroscopically measured temperature to the temperatures measured with thermocouples. In this study, a calibration curve for the intensity ratio of the two Swan bands vs the thermocouple temperature was constructed from the simultaneous measurements of the two using a premixed propane-air flame mixture in the test furnace. The temperature was changed by altering the equivalence ratio and the flow rates of gases. Figure 3 shows the calibration curve obtained with these measurements. The intensity ratio of the two Swan bands, observed at each pixel, was converted to the thermocouple temperature, which was then used to construct the temperature profile.

Results and Discussion

Figure 4 shows the two-dimensional distribution of temperatures in the flame measured with the thermocouple array for the three different flame conditions examined here. These conditions include 1) air having normal oxygen concentration of 21% at room temperature, 2) normal oxygen concentration of 21% in air heated to a temperature of 1200 K, and 3) oxygen concentration of 4% in air (called diluted air) at high temperature. Figure 4 shows the results on temperature distribution in the combustion chamber for the three cases. These results show that the distribution of temperatures with the ordinary air, containing 21% oxygen concentration in air, for both the heated and normal temperature air, are similar (compare Figs. 4 conditions 1 and 2). In both cases, steep temperature gradient can be found. The peak temperature for the heated air case with normal oxygen concentration air is much higher than the unheated normal temperature air case.

In contrast, the global flame temperature with high temperature and low oxygen concentration air is lower than that found for the preceding two cases with the normal oxygen concentration air. The thermal field uniformity is significantly enhanced with highly preheated and low oxygen concentration (diluted) air at high air-preheat temperature (see Fig. 4 condition 3).

Figure 5 shows the spatial distribution of emission intensity of combustion radicals, such as, C₂, CH, and OH in the two flames at two different oxygen concentrations in air. These measurements were made with the UV-CCD camera having an interference filter that could only pass the light signal for the specie to be detected. Condition 1 is for normal 21% air, whereas condition 2 is for diluted 4% oxygen concentration air preheated to 1200 K. The profile measured with the 1200-K air-preheat temperature and 21% oxygen

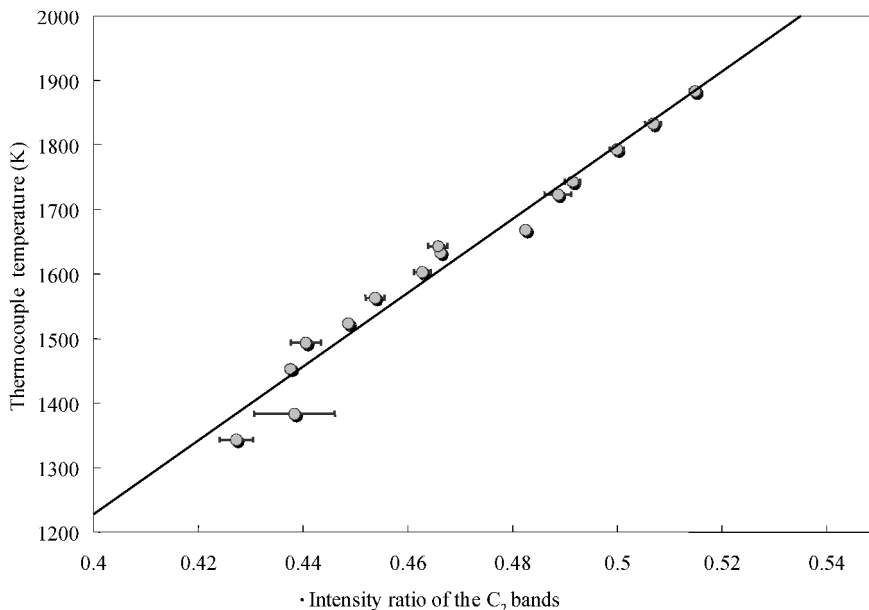


Fig. 3 Calibration curve for temperature calculated from the C_2 emission intensity ratio.

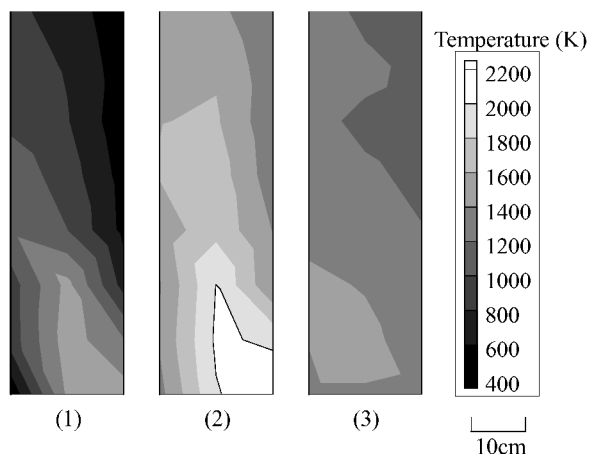


Fig. 4 Measured temperature profiles obtained with the thermocouple array: condition 1, air temperature = 300 K, oxygen concentration in the air = 21 %; condition 2, air temperature = 1200 K, oxygen concentration in the air = 21 %; and condition 3, air temperature = 1200 K, oxygen concentration in the air = 4 %.

concentration in the air is not shown in Figs. 5 because of the very high background emission levels, which prevented the detection of emission signal from the radicals. Note that the observed emission intensity levels for condition 2, using highly preheated and low oxygen concentration air, are much lower for all of the species measured as compared to those measured for the normal air condition 1. Detailed comparison of conditions 1 and 2 (Fig. 5) reveals that, under condition 1, the CH emission intensity level (Fig. 5b) is almost at the same level as the C_2 emission level (Fig. 5d), but higher than the OH emission levels (Fig. 5a); compare the maximum levels. In contrast, for condition 2, the CH emission level is lower than both the OH and C_2 emission intensity levels (see condition 2, Figs. 5a and 5d). The reasons for these observed phenomena are considered to be as follows. The spectral intensity for the electronic transition, or the integrated intensity including all of the related rotational and vibrational transitions, changes depending on the flame temperature T and the number of combustion radicals N . The intensity can be expressed as

$$I = h\nu NgA \exp(-E/kT)/Q(T) \quad (2)$$

where h is the Planck's constant, ν the frequency, N the number density, g the statistical weight of the excitation level, A the transition probability for spontaneous emission, E the upper level energy, k the Boltzmann's constant, $Q(T)$ the partition function. The first phenomenon can be explained as follows. A slight decrease in flame temperature between the two methods can be seen in the central region for condition 2. Note that the spectroscopically measured temperature profiles provides high spatial resolution as seen from the comparison between Fig. 6 conditions 1 and 2. This is not clear with the temperature measured with the thermocouples. As can be seen from the exponential term in Eq. (2), a slight decrease in temperature leads to significant decrease in emission intensity from all of the species unless the number density is increased drastically.

For the comprehension of the second phenomenon, it is useful to take a ratio of the emission intensity for the two species, that is,

$$I_{C_2}/I_{CH} = K_1(N_{C_2}/N_{CH}) \exp(E_{CH} - E_{C_2})/kT \quad (3)$$

Similarly,

$$I_{OH}/I_{CH} = K_2(N_{OH}/N_{CH}) \exp(E_{CH} - E_{OH})/kT \quad (4)$$

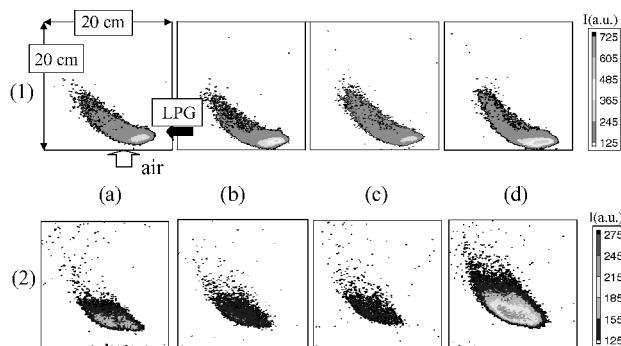


Fig. 5 Emission intensity profiles of OH, CH, and C_2 radicals in the flames: condition 1, air temperature = 300 K, oxygen concentration in the air = 21 % and condition 2, air temperature = 1200 K, oxygen concentration in the air = 4 %; a) OH (306 nm), b) CH (431 nm), c) background emission (480 nm), and d) C_2 (515 nm).

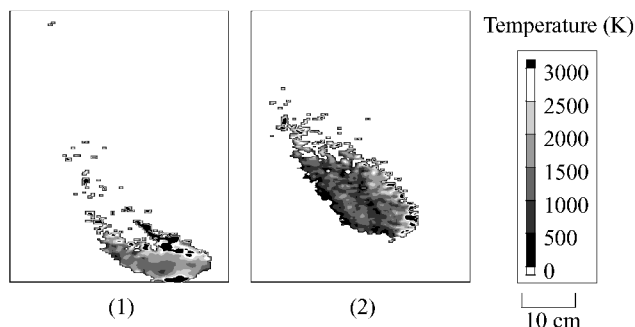


Fig. 6 Temperature profiles calculated from the ratio of C_2 emission intensities with the calibration curve: condition 1, air temperature = 300 K, oxygen concentration in the air = 21% and condition 2, air temperature = 1200 K, oxygen concentration in the air = 4%.

where the constants K_1 and K_2 involve the ratios of the term $v_g A/Q(T)$ [$Q(T)$ and their value is almost constant at temperatures below 2500 K]. The difference in the excitation energy in the exponential term is either positive [Eq. (3)] or negative [Eq. (4)],⁹ because $E_{CH} = 23150 \text{ cm}^{-1}$, $E_{C_2} = 19306 \text{ cm}^{-1}$, and $E_{OH} = 32682 \text{ cm}^{-1}$. Consequently, the exponential term increases with the decrease in the temperature in Eq. (3), whereas it decreases in Eq. (4). The relatively low intensity of CH emission as compared to the C_2 emission for the case of highly preheated air with the low oxygen concentration condition 2 is consistent with the hypothesis given earlier. In contrast, it is suggested that by taking into account the increase in I_{OH}/I_{CH} and the decrease in the exponential term, the number density of OH radicals become relatively large as compared to the CH radicals. This might be related to slow combustion under condition 2. To determine the number density of radical species, further investigation must be made using some other spectroscopic method, such as laser-induced fluorescence diagnostics.

The flame temperature can also be determined from the ratio of C_2 emission intensities obtained at two (471 and 515 nm) wavelengths. The detailed theory for the calculation, based on Eq. (2) for the modified two-line method or two-band method, has been described in our previous study.⁶ In this study, the relationship between the temperature measured with a thermocouple and the spectral intensity ratio of the C_2 bands that reflects the vibrational temperature was determined experimentally (see the calibration curve shown in Fig. 3). The calibration curve was made by measuring the thermocouple temperature and the spectral intensities at a same point in a premixed propane/air flame supported on a slot burner. When this calibration curve was used, the spectral intensity ratios at the CCD pixels were converted into the thermocouple-equivalent temperature, which then allowed determination of the temperature profiles.

The profiles for the net emission intensity of C_2 radical have been estimated by subtracting the background intensity level (at 490 nm) from C_2 emission intensity level (at 471 or 515 nm). Figure 6 shows the calculated temperature profiles obtained for the two cases of normal air at room temperature and highly preheated air having low oxygen concentration. The results show detailed structure of the thermal field as compared to that obtained with the thermocouple array (compare the results shown in Fig. 6 with those shown in Fig. 4.) The temperature profiles obtained with the Swan band indicate only the flame region or the combustion reaction zone where the radiation from C_2 radicals exists. This also clearly demonstrates the flame front region. The two figures reveal that the temperature gradient becomes smaller with diluted air combustion.

Figure 7 shows the two-dimensional distribution of flame fluctuation for the three combustion conditions that have been obtained using Eq. (2). Note that highly preheated air has a significant effect on decreasing the flame fluctuations in the main (middle) combustion region, compare condition 1, Fig. 7, with conditions 2 and 3, Fig. 7. The practical importance of the highly preheated air combustion is, therefore, very significant from the point of view of both thermal field uniformity and flame stability. Similar trends on the profiles of the temperature fluctuation has also been obtained with

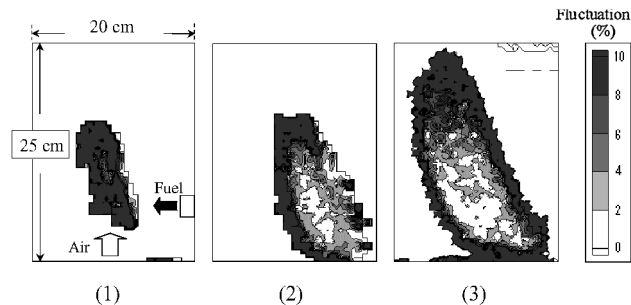


Fig. 7 Profiles of flame fluctuation obtained with the high-speed video camera and FFT: condition 1, air temperature = 300 K, oxygen concentration in the air = 21%; condition 2, air temperature = 1200 K, oxygen concentration in the air = 21%; and condition 3, air temperature = 1200 K, oxygen concentration in the air = 4%.

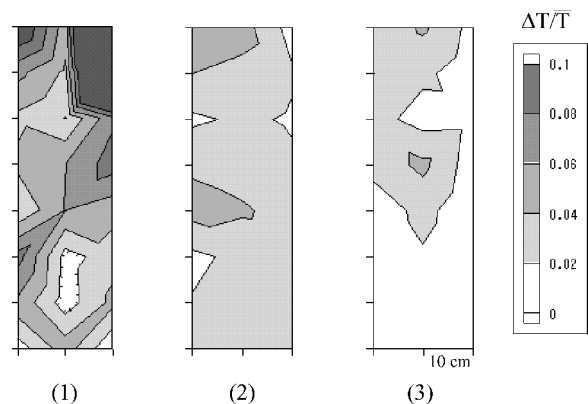


Fig. 8 Profiles of flame temperature fluctuation obtained with the high-speed video camera and FFT; flame conditions are the same as given for Fig. 7.

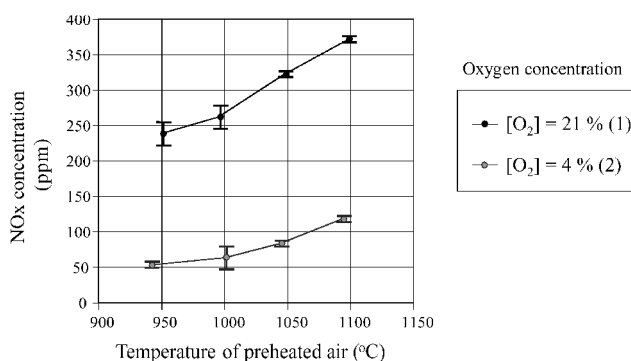


Fig. 9 Effect of diluting O_2 concentration in the air on NO_x emission: condition 1, oxygen concentration in the air = 21% and condition 2, oxygen concentration in the air = 4%.

the data obtained using thermocouples followed by the FFT procedure (see Fig. 8).

Figure 9 shows the NO_x emission levels as a function of temperature of the highly preheated combustion air. The NO_x concentration in the exhaust gas vent was measured with a chemiluminescence NO_x analyzer using a probe sampling technique. The results presented in Fig. 9 clearly show that the NO_x emission level drastically decreases at any air preheat temperature by diluting the preheated combustion air. Thus, it is possible to achieve simultaneously low NO_x emission, high thermal field uniformity, and low flame fluctuations. Low fluctuations of the flame mean uniform heating of the object being heated with the flame. Furthermore, significant energy saving through the use of heat regenerators is also concurrently achievable.^{4,5}

Conclusions

The results presented here with the thermocouple array and the UV-CCD camera have shown that the use of low oxygen concentration and high-temperature air significantly enhances the flame thermal uniformity of flames. The use of highly preheated low oxygen concentration air resulted in larger combustion volume, wider (broader) spatial distributions of emission intensity from the combustion radicals, and lower peak flame temperatures. The profiles of spontaneous emission intensity from combustion radicals provide information on the temperature and number density of these radicals. The results obtained from the observation of the high-speed video camera have shown good flame stability with low oxygen concentration air at high degrees of air preheat. The flame fluctuations were reduced with the use of high-temperature and low oxygen concentration air. Low peak flame temperatures are instrumental for reducing NO_x emission. The high-temperature and low oxygen concentration combustion air resulted in low NO_x emission levels.

Acknowledgments

The support provided by Nippon Furnace Kogyo (NFK) and NEDO Japan is greatly appreciated. The National Science Foundation NASA also provided support to this research to Ashwani K. Gupta and is gratefully acknowledged. He would also like to thank Toshiaki Hasegawa, of NFK for all his support and contributions to this effort.

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