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Temperature Dependence of Intensities of Laser-Induced Fluorescences of Ethylbenzene and Naphthalene Seeded in Gas Flow at Atmospheric Pressure

Implications for Quantitative Visualization of Gas Temperature

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Abstract : The present study has been carried out with the aim of developing a technique for measuring two-dimensional gas temperature profiles based on two-color fluorescence induced by a one-color laser. The laser sheet of the fourth harmonic (266 nm) from a Nd:YAG laser induced fluorescence in species doped in a nitrogen gas flow. The LIF spectra of seven fluorescent species, namely acetone, methylethylketone, acetaldehyde ethylbenzene, anisole, aniline, and naphthalene, were measured to select the best prospective pair of fluorescent species for this technique. Ethylbenzene and naphthalene show relatively high LIF intensities and their LIF spectra overlap less with each other than with other species. Also, ethylbenzene has a high temperature dependence while naphthalene has a low temperature dependence. Thus by selecting one portion of wavelengths in the range where ethylbenzene or naphthalene is dominant, the temperature of the gas can be determined using the ratio of LIF intensities of the mixture at the two wavelengths with good temperature sensitivity. In addition, a general principle is presented for finding out an optimum pair of wavelengths to obtain a good temperature sensitivity in those LIF spectra.

Keywords : Temperature Measurement, Laser Induced Fluorescence, Fluorescent Species, Temperature Sensitivity, Laser Sheet.

1. Introduction

The laser-induced fluorescence (LIF) technique has been widely applied for diagnostics on reactive fluid systems. Spectral distributions and/or intensities of LIF from species in the fluids may yield their velocity, density, temperature and pressure. If temporal developments of two-dimensional distributions of these quantities are obtained, useful studies such as fuel mixing and evaporation processes for combustion investigation can be carried out.

If species of the fluids for study are not suitable for LIF, tracers are to be seeded into the fluid to carry out the measurement. For the latter case, selection of suitable tracers for the purpose of the measurement at hand using LIF is of utmost importance. For example, acetone is one of the preferred tracers for visualization of fuel mixing and evaporation processes because it does not pyrolyze significantly up to 900 K, if the residence time is not too long (Thurber et al., 1998). However, its evaporation characteristics differ too much from those of isooctane, which is used as a standard liquid fuel in combustion engine experiments. The ideal tracer should behave exactly like the fluid to which it is added (Schulz and Sick, 2005). Since evaporation characteristics of 3-pentanone are very similar to those of isooctane, 3-pentanone is considered a better tracer than acetone to be seeded into the liquid fuel. As tracers for the visualizations of fuel mixing and evaporation processes, an exciplex of TMPD and naphthalene is a prospective candidate because separate measurements of LIF from injected liquid fuel and its vapor may be possible (Melton, 1983). The advantage of the different responses of 3-pentanone and toluene to collisional quenching by molecular oxygen was used for the measurement of oxygen concentration (Koban et al., 2002).

The present study aims at developing a technique to measure two-dimensional gas temperature profiles based on two-color fluorescence detection from two species induced by a one-color laser. The technique based on a similar concept has been applied several times to liquid (Sakakibara et al., 1999; Funatani et al., 2004). Recently, Baranger et al. (2005) reported dependencies of the LIF spectrum and intensity of kerosene on temperature, pressure, and oxygen mole fraction. They showed that the temperature dependences of one- and two-ring aromatic species were different but their main interest was in visualization of oxygen mole fraction rather than the temperature measurement. Thus, the investigation is still limited and has not been established yet.

The present technique would be limited to isobaric conditions because the pressure dependences of two fluorescent species would not cancel out. Thus the potential application of the present technique may be measurements of temperature-field of open thermal systems in the range of around room temperature or higher, e.g. temperature-field around heat transfer devices, such as in an automobile engine chamber or in an air-conditioner system.

Making quantitative visualization of temperature-fields using LIF techniques has encountered significant difficulties. One of the reasons for these difficulties is that the intensity of laser induced fluorescence depends not only on the temperature but also on the number density and optical characteristics of a fluorescent species as well as the laser intensity. The laser intensity distribution in a laser sheet is usually not uniform, due to absorption or dispersion by gas molecules in addition to the intensity distribution of the laser sheet itself, which leads to a measurement error in the gas temperature. One of the primary aims of the present study is to try to reduce this error. In the present measurement technique, the temperature of the gas is to be determined using the ratio of LIF intensities at the two wavelengths from the mixture of the two fluorescent species. Thus, one of the merits of this technique is that the influence of the laser intensity distribution on a sheet is cancelled out since the laser light is emitted from a single laser system.

In choosing the excitation wavelength, special attention was paid on the possibility to form a wide laser sheet of high intensity without excessive cost. For this purpose, we focused on 266 nm which is emitted as the fourth harmonic of a Nd:YAG laser. At the first stage of the development, it was necessary to select a pair of fluorescent species and fluorescence wavelengths. Fluorescent species were selected on the premise that they are excited at 266 nm and not too explosive and not toxic to humans. By bearing these in mind, LIF spectra of the seven fluorescent species in nitrogen gas flow were then measured. Temperature dependences of LIF spectra were measured first to select an optimum pair of fluorescent species and then an optimum pair of fluorescent wavelengths for the selected species. For these selections, a general principle was established for finding out an optimum pair of wavelengths to obtain a good temperature sensitivity in those LIF spectra.

2. Experimental Setup and Method

Figure 1 shows an optical layout of an excitation light source. A laser pulse of the second harmonics

(532 nm) of an Nd:YAG laser (The New Wave Research, Solo 120) lasting for 3 to 5 ns at a repetition of 10 Hz was emitted through a SHG (second harmonic generator) installed in the laser unit. This second harmonics light went through a SHG outside the unit and then a part of it became the fourth harmonics (266 nm). In order to remove the second harmonics, four mirrors were placed in the optical path which reflected the fourth harmonics but transmitted the second harmonics. The overall intensity of the laser beam at the fourth harmonics was 7 mJ. The laser beam then went through a cylindrical lens and a converging lens to form a laser sheet having a width of 12 mm and a thickness of 1.0 mm.

The schematic drawing of the gas flow and measurement system is shown in Fig. 2(a). In the present study, we have focused on developing a technique to measure temperature as accurately as possible. We intend to use nitrogen as a carrier gas of the tracers for the temperature measurement, thus have excluded the species that would cause severe quenching, e.g. oxygen. Nitrogen gas in a cylinder was sent, at a regulated pressure, to a mass flow controller (Horiba STEC, SEC-E40MK3) which controlled the volume flow rate at the standard state of the nitrogen gas at 4.0 L / min. The gas was doped with a liquid fluorescent species by bubbling in a doping chamber, as shown in Fig. 2(b), or doped with a solid fluorescent species by going through a solid fluorescent species wrapped in a sheet of breathable paper which was installed in a doping chamber, as shown in Fig. 2(c). Both doping chambers were located in water to keep them at a roughly constant temperature (290 K). Those fluorescent species were exposed to nitrogen gas sufficiently long to attain a saturated vapor pressure at the temperature.

After the doping chamber, the doped gas was filtrated to remove liquid or solid fluorescent species, as shown in Fig. 2(a) again. Then, it flew between two concentric tubes, the outer tube of which was surrounded, from the inside to outside, with a ceramic tape, kanthal wires, and a thermal insulant. Air flow was kept at a controlled flow rate, heated by a gas heater and then sent inside of the inner tube. In this way, the doped gas flow was heated up from both inner and outer tubes up to a specific temperature. The heated doped gas passed through a wire mesh flow-straightener and then was sent to a quartz cell for optical measurement. At the other end of the quartz cell, a port was placed to exhaust the gas.

The laser sheet was irradiated into the center of the quartz cell through a slit and absorbed by fluorescent species. The laser sheet was finally sent to a narrow rectangular piece of UV absorption filter in the tube, which acted as a beam dump. A collective lens having a diameter of 5.0 mm and a focal length of 25.2 mm, and a receiver lens having a diameter of 5.0 mm which was directly attached to the optical fiber end in a housing, were perpendicularly placed to the axis of the laser sheet. The optical fiber was connected to a spectroscope (Lambda Vision, SA-100HPCB) for the measurement of the fluorescent spectra. The exposure time of spectroscope was 2.0 s for all experiments.

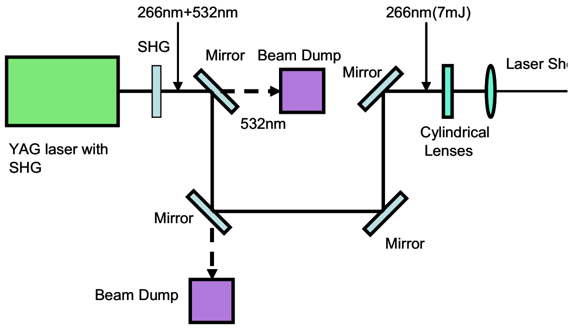

Fig. 1. Optics layout.

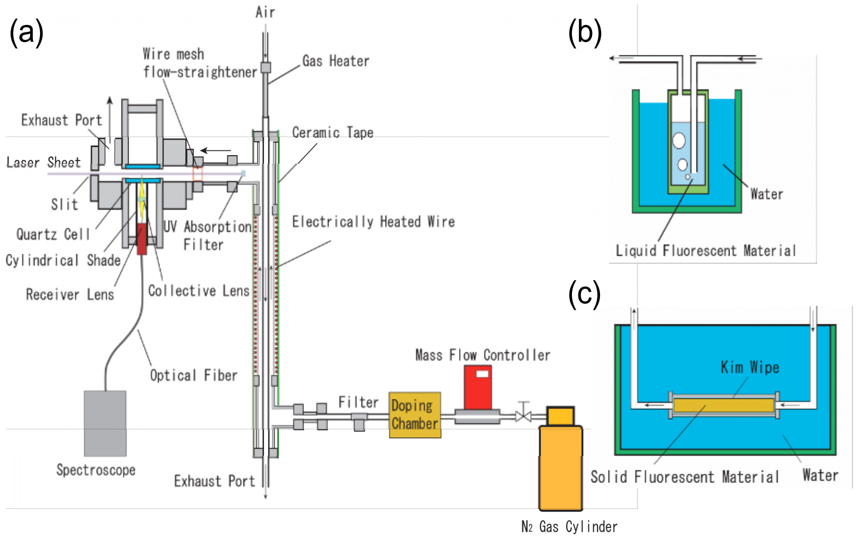

Fig. 2. Schematic drawing of the experimental arrangement:(a) the piping layout and measurement system. (b) the doping chamber for ethylbenzene. (c) the doping chamber for naphthalene.

Just prior to the spectra measurement, temperature of gas flow at the focus point of spectroscope was measured using a type K thermocouple having a diameter of ϕ1.6 mm which was inserted through the slit.

3. Experimental Result and Discussion

The cross-sectional average flow velocity in a quartz cell was 0.667 m/s at 273 K, which increased with increasing gas temperature. It should be noted that the thermal expansion of gas, due to the increase in temperature, decreases the number density of fluorescent molecules. The present study intends to measure gas temperature at an atmospheric pressure. Thus, the effect of temperature on LIF intensity is later discussed, including the effect of the temperature on the number density of fluorescent molecules.

Seven prospective fluorescent species at 295 K were induced by a laser sheet at the wavelength of 266 nm and the resultant fluorescent spectra were measured. The LIF spectra of acetone, methylethylketone and acetaldehyde are shown in Fig. 3(a), and those of ethylbenzene, anisole, aniline and naphthalene are shown in Fig. 3(b), all at room temperature. The LIF spectra of fluorescent species shown in Fig. 3(a) are broadly distributed from 300 nm to 650 nm with relatively low intensities. The LIF spectra of fluorescent species shown in Fig. 3(b) are distributed in narrower ranges of wavelength with much higher intensities than those shown in Fig. 3(a). The LIF spectrum of anisole shows the highest peak in those fluorescent species. However, its spectrum overlaps the spectra of the other three fluorescent species shown in Fig. 3(b). On the other hand, the spectral distributions of ethylbenzene and naphthalene overlap each other less and have relatively high LIF intensities. Thus, we selected ethylbenzene and naphthalene as one of prospective pair of fluorescent species from these seven fluorescent species and show them again in Fig. 4. From this figure, it can be seen that the LIF intensity in the range of 250 nm to 315 nm comes mainly from ethylbenzene and that in the range of 315 nm to 410 nm does mostly from naphthalene.

The next concern is whether the temperature dependences of these species are sufficient for the purpose of temperature measurements. The LIF spectra of ethylbenzene at various temperatures are shown in Fig. 5(a-1) and their normalized forms relative to the LIF spectrum at 295 K in Fig. 5(a-2). Similar figures for naphthalene are shown in Figs. 5(b-1) and 5(b-2). The wavelengths for the peak intensities of ethylbenzene and naphthalene showed a shift in no more than few nm to longer wavelengths. In addition, the range of LIF spectra showed no obvious shifts.

The LIF intensities of both species decreased as the temperature was increased. While the gas temperature was increased from 295 K to 356 K, the overall LIF spectrum of ethylbenzene decreased about 60 % and that of naphthalene 30 %. Thus, the temperature dependences of ethylbenzene and naphthalene are significantly different.

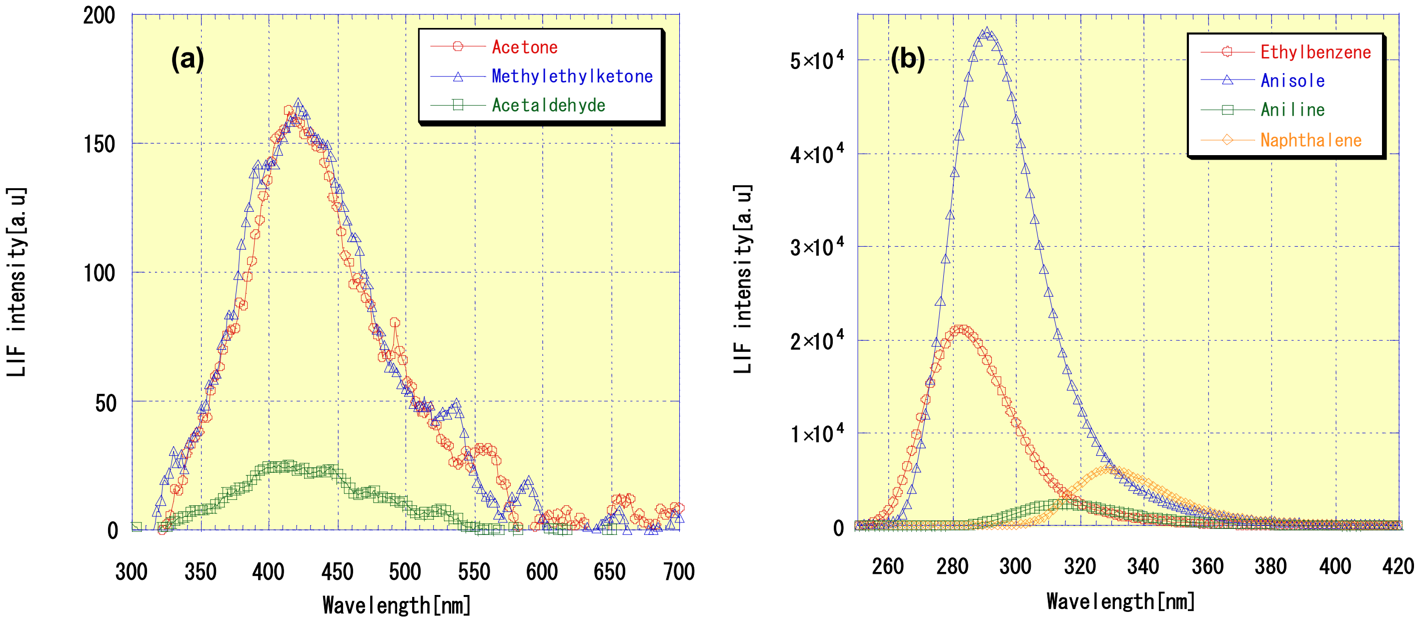

Fig. 3. LIF spectra of fluorescent species induced by a laser sheet at the wavelength of 266 nm.

In the expression of fluorescence intensity, there are three temperature-dependent terms, which are the number density, the molecular absorption cross section of the fluorescent species, and the fluorescence quantum yield (Thurber et al., 1998). Number densities of fluorescent species decrease as temperature is increased due to thermal expansion. Because number densities of both species change in the same ratio as temperature changes, we conclude that the above observation of different temperature dependences is due to the molecular absorption cross section of the species and the fluorescence quantum yield in the mentioned three terms.

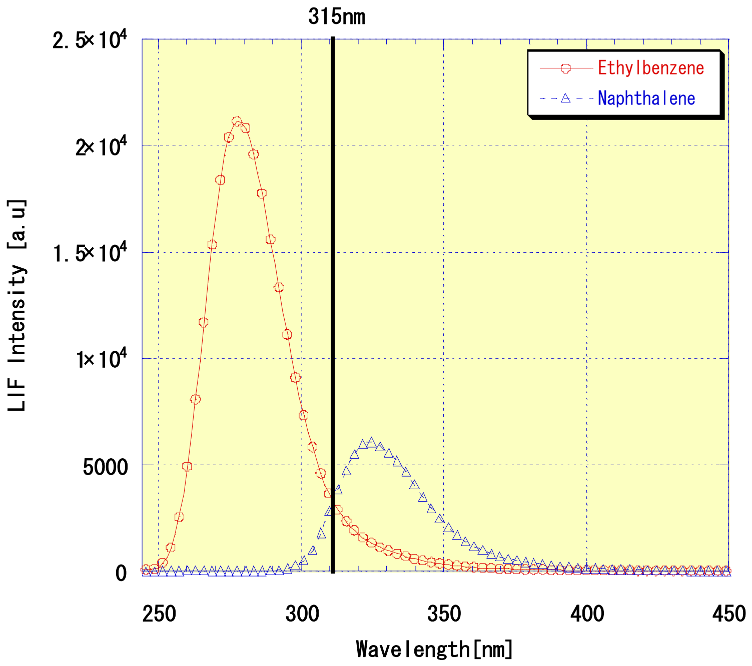

Fig. 4. LIF spectra of ethylbenzene and naphthalene.

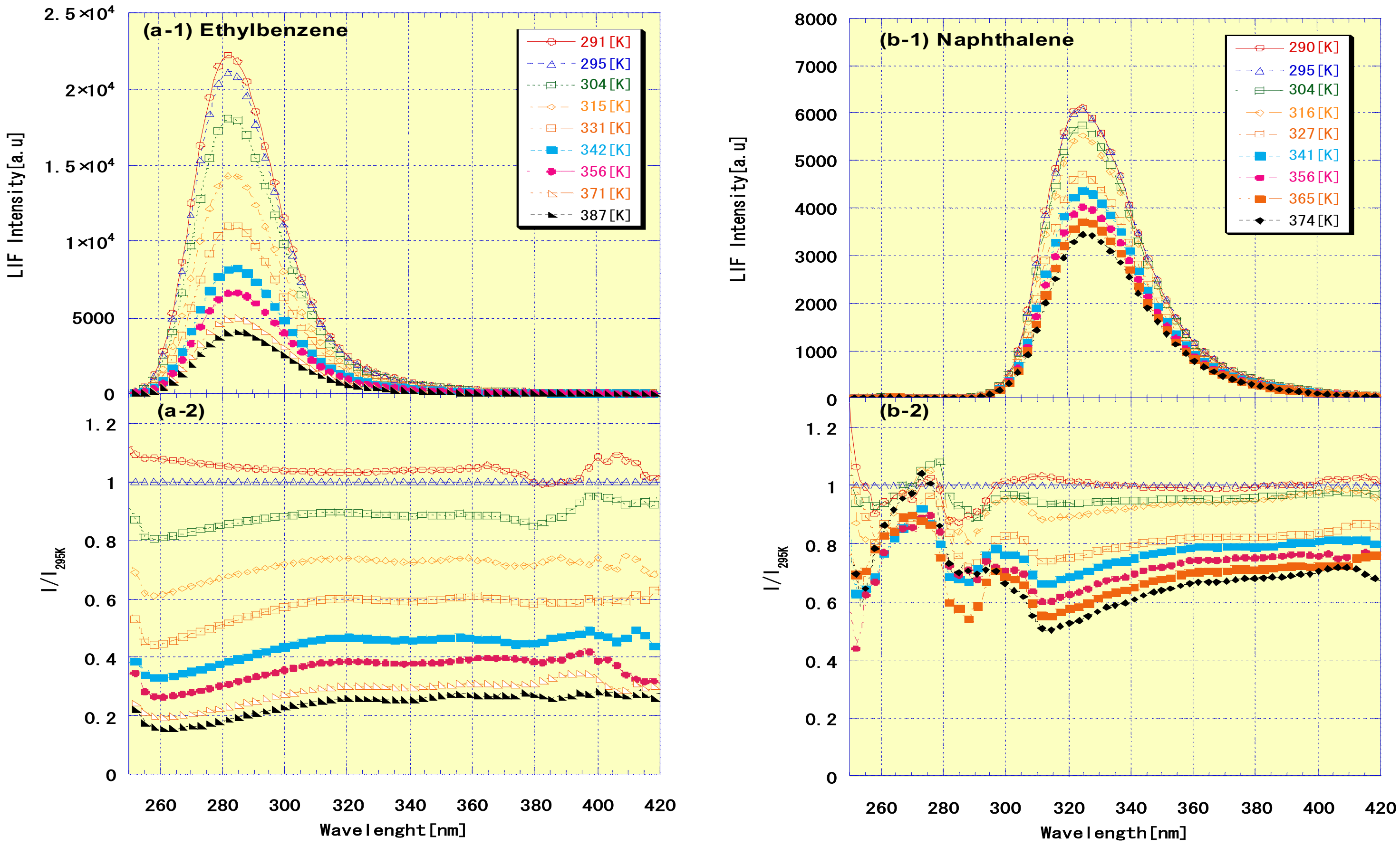

Fig. 5. LIF Spectra at various temperature: (a-1) and (a-2) are for ethylbenzene; (b-1) and (b-2) are for naphthalene; (a-2) and (b-2) are normalized forms relative to the LIF spectra at 295 K.

Figures 5(a-2) and 5(b-2) show that the largest ratio of the LIF intensity decrease is attained at around 260 nm for ethylbenzene and at around 315 nm for naphthalene. It should be noted that both these wavelengths are shorter than the wavelengths at which the LIFs show the peak intensities. It may be of interest to further note that this trend is similar to that of the spectra of thermal emission.

Next, we discuss the strategy for selecting the best pair of wavelengths for these fluorescent species, which are denoted by λ_S and λ_l. The factors which have to be taken into accounts are (i) the rate-of-change of the ratio of LIF intensities at the two wavelengths, by the temperature difference, namely $I(\lambda_s, T)/I(\lambda_l, T)$, where the function I indicates the LIF intensity, and (ii) the intensity of LIF itself which decides the S/N ratio of the measurement.

Regarding the point (i) above, we first discuss the case when there is a single fluorescent species. In order to more clearly see trends of the LIF intensities and the normalized LIF intensities on temperature, Figs. 6(a-1), 6(a-2), 6(b-1) and 6(b-2) illustrate these quantities as a function of temperature. Figures 6(a-1) and 6(a-2) are for ethylbenzene in the range of 254 to 315 nm. Similarly, Figs. 6(b-1) and 6(b-2) are for naphthalene in the range of 315 to 412 nm. Although the change of LIF intensities due to the temperature difference from 290 K to 370 K is, as shown in Fig. 6(a-1), the largest at 285 nm which is close to the fluorescence peak wavelength, the rate of change of LIF intensities are more important as a measure of temperature sensitivities. For this purpose, Fig. 6(a-2) illustrates the normalized LIF intensity against that at 295 K. This figure shows the optimum wavelength for ethylbenzene to be 264 nm. Similarly, the optimum wavelength for naphthalene is 315 nm from Fig. 6(b-2).

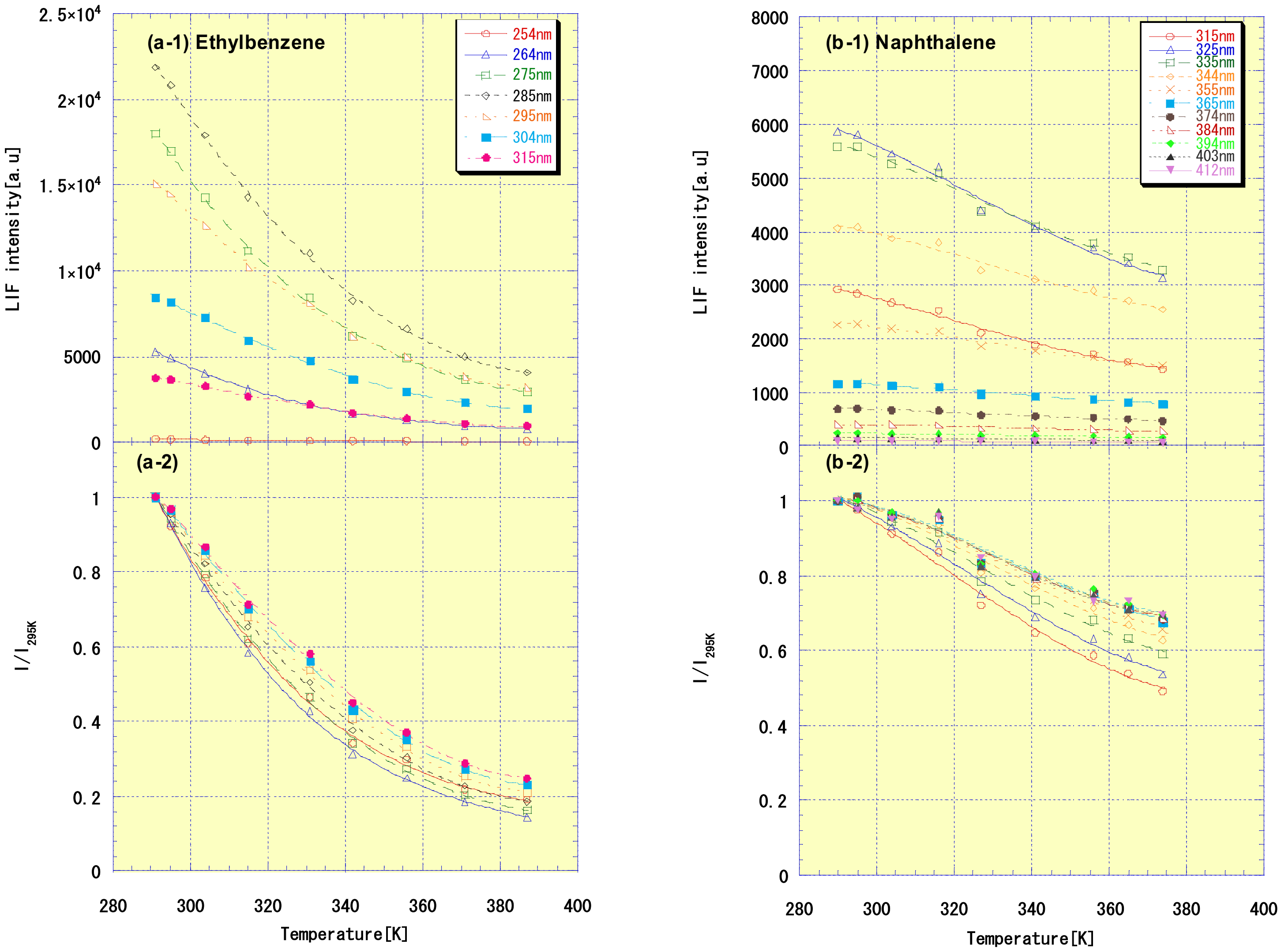

Fig. 6. LIF intensities as a function of temperature: (a-1) and (a-2) are for ethylbenzene; (b-1) and (b-2) are for naphthalene; (a-2) and (b-2) are normalized forms relative to the maximum LIF intensities in each wavelength.

Then, we discuss the case when the two fluorescent species are mixed into a gas flow. In the present technique, we modeled the LIF intensity ratio of the mixture of these florescent species to estimate temperature sensitivity, assuming the quenching effect be negligible. This assumption is supported by the fact that the quenching effect is nearly constant due to the constant ratio of concentration of both fluorescent species.

We define a function R as $\mathrm{R}(\lambda_s,\lambda_l,T)\equiv \mathrm{I}(\lambda_s,T)/\mathrm{I}(\lambda_l,T)$. The ratio of peak intensities of LIF spectra of both species depends on the species loading and, thus, the ratio is arbitrary. Therefore, we make the ratio unity by normalizing them by their peak intensities. If we mix the fluorescent species to have the same peak intensities, $\mathrm{R}(\lambda_s, \lambda_l, T)$ becomes

$$\mathrm{R}(\lambda_s,\lambda_l,T)=\frac{\mathrm{I}_e(\lambda_s,T)/\mathrm{I}_{e0}+\mathrm{I}_n(\lambda_s,T)/\mathrm{I}_{n0}}{\mathrm{I}_e(\lambda_l,T)/\mathrm{I}_{e0}+\mathrm{I}_n(\lambda_l,T)/\mathrm{I}_{n0}} \tag{1}$$

where I_{e0} and I_{n0} are the measured peak intensities, and the subscript e and n denote ethylbenzene and naphthalene, respectively.

The rate-of-change of $\mathrm{R}(\lambda_s, \lambda_l, T)$ gives the temperature sensitivity of the present technique between T_1 and T_2, namely

$$\Delta\mathrm{R}^*(\lambda_s,\lambda_l,T_1,T_2)\equiv\frac{\mathrm{R}(\lambda_s,\lambda_l,T_2)-\mathrm{R}(\lambda_s,\lambda_l,T_1)}{\mathrm{R}(\lambda_s,\lambda_l,T_1)} \tag{2}$$

The combination of λ_s and λ_l is to be sought using values of $\Delta\mathrm{R}^*(\lambda_s, \lambda_l, T_1, T_2)$ to obtain the optimum temperature sensitivity at a given temperature range.

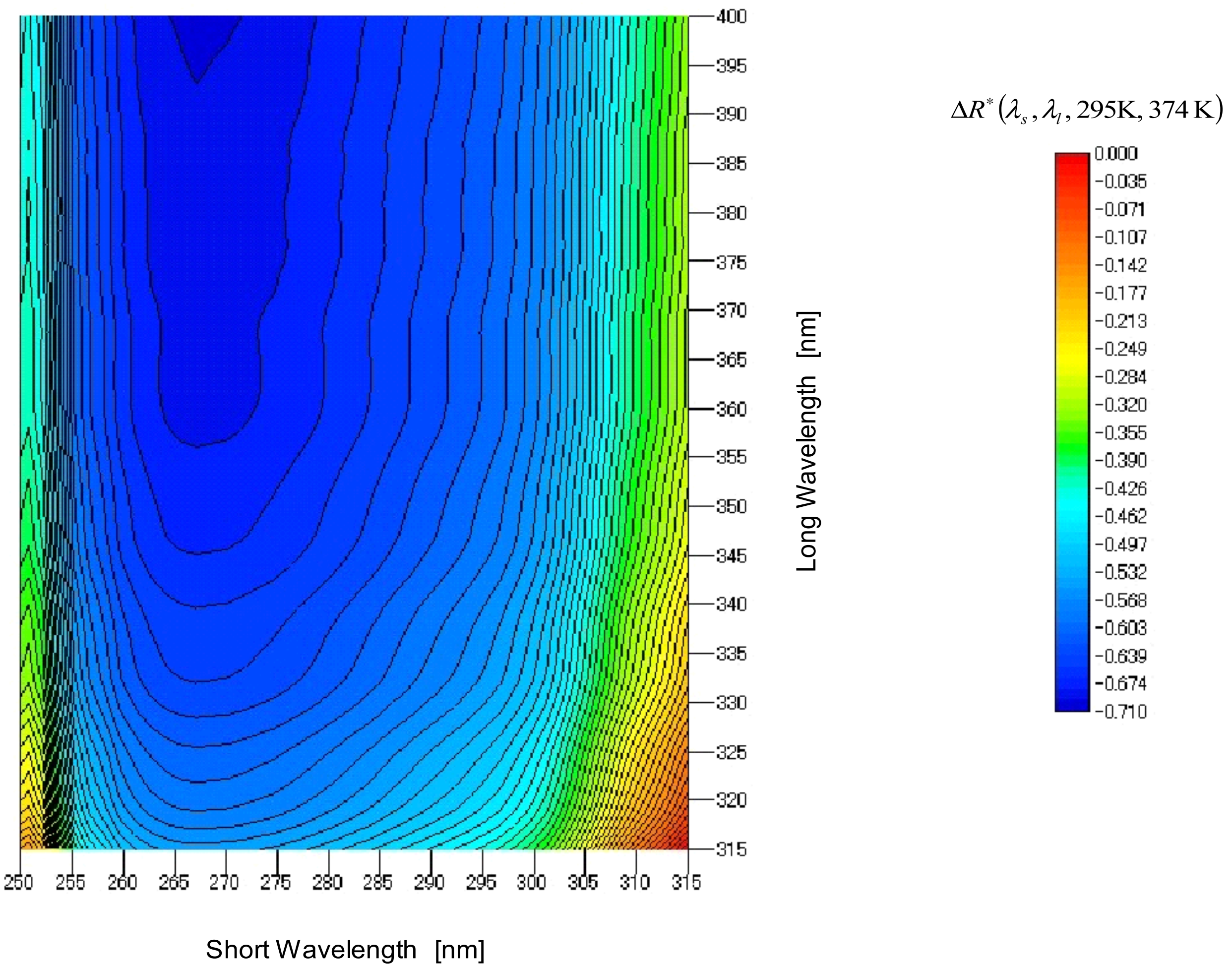

Fig. 7. Equi-contour plots of $\Delta\mathrm{R}^*(\lambda_s, \lambda_l, T_1, T_2)$ for T_1 = 295 K and T_2 = 374 K, at various values of λ_s and λ_l.

Figure 7 shows equi-contour plots of $\Delta R^*(\lambda_s, \lambda_l, 295\ \mathrm{K}, 374\ \mathrm{K})$, namely the rate-of-change of $R(\lambda_s, \lambda_l, T)$ at the temperatures of $T_1 = 295$ K against $T_2 = 374$ K, at various values of λ_s and λ_l. From this figure, the optimum wavelengths in the sense of temperature sensitivity were found to be $\lambda_s = 267$ nm and $\lambda_l = 400$ nm, respectively, where $\Delta R^*(\lambda_s, \lambda_l, 295\ \mathrm{K}, 374\ \mathrm{K})$ attained about 70 %. Because shapes of equi-contour plots for different values of T_2 do not change much from those shown in Fig. 7, as may be imagined from Figs. 5(a-2) and 5(b-2), these optimum wavelengths hold for other values of T_2.

Regarding the second point (ii) above, we discuss the choice of 267 nm and 400 nm from the viewpoint of an S/N ratio of the measurement. Since the wavelength of 267 nm is close to the shorter edge of the LIF spectra of ethylbenzene as shown in Fig 5(a-1), it may be necessary to select a longer wavelength than this, if an S/N ratio is not sufficient. Also, we see that 400 nm is exactly at the longer edge of the LIF spectrum, as can be seen from Fig. 5(b-1). Thus, this latter wavelength may also be determined in practice as long a wavelength as possible as far as an S/N ratio of the measurement allows.

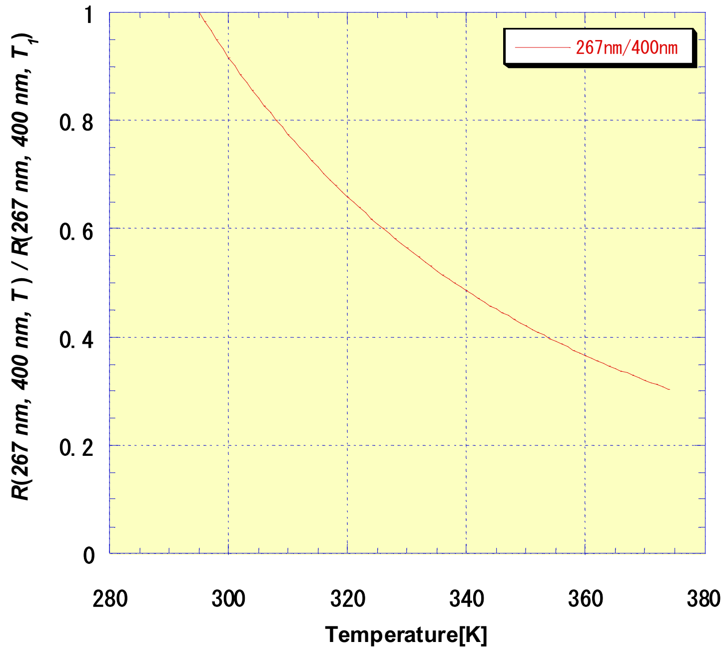

Fig. 8. Temperature-intensity ratio characteristic curve, namely the signal ratio of the LIF intensities at 267 nm and 400 nm against temperature.

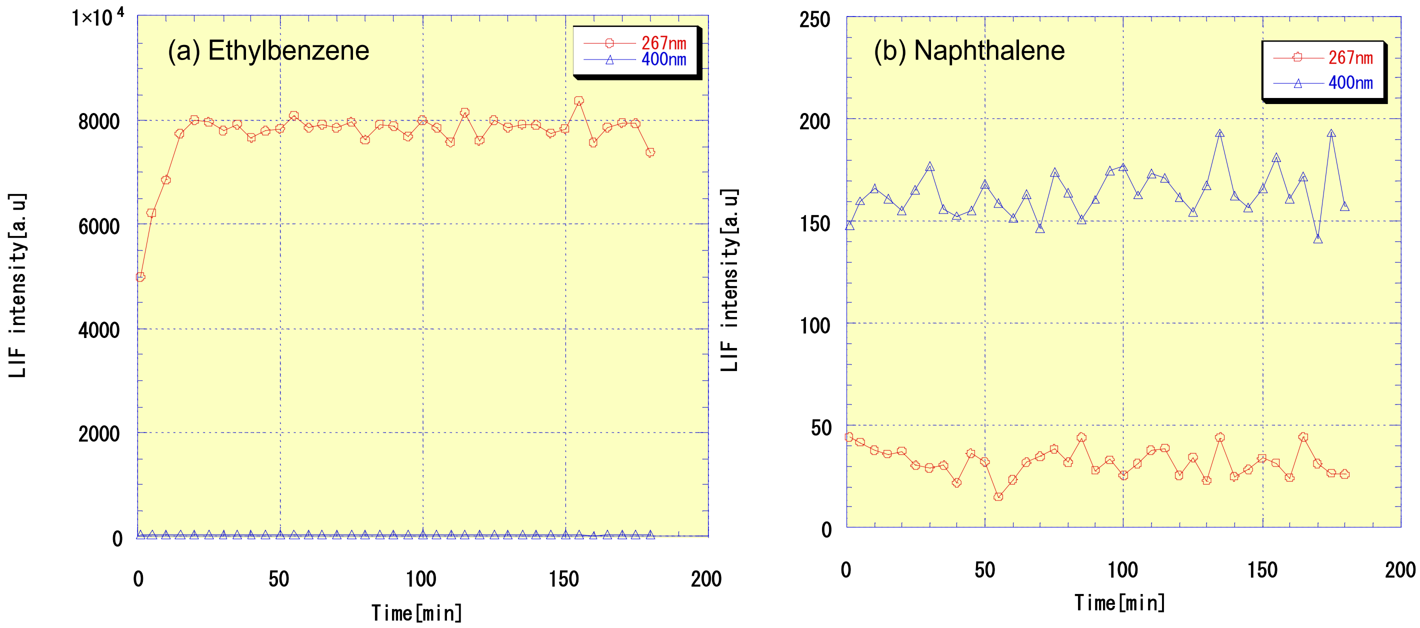

Fig. 9. Temporal changes of LIF intensities of (a) ethylbenzene and (b) naphthalene at 267 nm and 400 nm.

Once the optimum pair of wavelengths has been determined, the next point is to obtain a temperature characteristic curve, namely the signal ratio of the LIF intensities at 267 nm and 400 nm against temperature. This is shown in Fig. 8. This characteristic curve monotonically decreases with increasing temperature of gas. This monotonic change ensures that a temperature is uniquely determined by this technique. The slope of the curve indicates the temperature sensitivity at each temperature. Thus, the curve indicates that the temperature sensitivity gradually worsens as the temperature increases. The temperature increase from 295 K to 374 K results in 70 % decrease in LIF intensity ratio from its value at 295 K. This means that the mean decrease of LIF intensity ratio per unit temperature increment is about 0.9 %. It is to be noted that this technique would be limited not only to the isobaric condition but also to relatively low temperatures because those LIF intensities and measurement accuracies decrease with increasing temperature.

In order to see the stability and reproducibility, thus to evaluate the measurement accuracy of the present technique, temporal changes of ethylbenzene and naphthalene LIF intensities at 267 nm and 400 nm were measured and are shown in Figs. 9(a) and 9(b), respectively. The LIF intensity of ethylbenzene became stable in 30 minutes after starting nitrogen flow. On the other hand, that of naphthalene had relatively large fluctuations from the beginning to the end. Fluctuations of measurement data are caused by the instability of species loading and laser intensity, and the optical, electrical and shot noises. The shot noise may be the main source of fluctuations in the present case, due to smaller LIF intensities for naphthalene, because other factors may be the same for both species.

Standard deviations of R obtained from each measurement data after 30 minutes is 7.1 % which corresponds to an accuracy of about 8 K. With regard to the point (ii) above, it is worthwhile to note that the best wavelength for the optimum temperature sensitivity may not be the same as the best wavelength for the optimum measurement accuracy. Depending on the instability or noise, the reconsideration of wavelength may be necessary to attain good accuracy. This topic will be discussed in the next study where two-dimensional temperature-field will be visualized using a CCD camera.

4. Conclusion

A laser sheet of the fourth harmonic (266 nm) of a Nd:YAG laser was applied to induce fluorescence from fluorescent species doped in a nitrogen gas flow. LIF spectra of seven fluorescent species namely, acetone, methylethylketone, acetaldehyde ethylbenzene, anisole, aniline, and naphthalene, were measured to select the best prospective pair of fluorescent species for this technique. Ethylbenzene and naphthalene show relatively high LIF intensities and overlap each other little. The spectral measurements at the gas temperatures of 290 K to 387 K revealed that this pair had a desirable characteristic, i.e. the spectra of ethylbenzene and naphthalene had different temperature dependences. This characteristic leads to good temperature sensitivity. Temperature is, in the proposed technique, to be determined by the ratio of LIF intensity of the mixture at two wavelengths. By selecting one of wavelengths in the range where ethylbenzene was dominant and another where naphthalene was dominant, good temperature sensitivity was attained.

These results indicate that ethylbenzene and naphthalene form a good pair of fluorescent species. Mapping the rate-of-change of the ratio of LIF intensities at various wavelengths is shown to be the way to find the optimum pair of wavelengths to obtain a good temperature sensitivity. It turned out that the optimum wavelengths are around the shorter edge of ethylbenzene spectra and on the longer edge of naphthalene spectra. These results show the feasibility of two-dimensional measurement of gas temperature based on two-color fluorescence induced by a one-color laser. Based on this technique, two-dimensional gas temperature profiles are to be studied as the next step.

Acknowledgements

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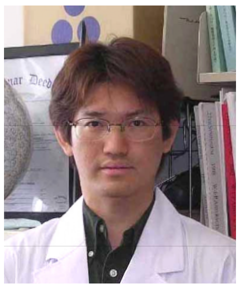

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