

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 -

Hirasawa, T. ^{*1}, Kaneba, T. ^{*2}, Kamata, Y. ^{*3}, Muraoka, K. ^{*1}, and Nakamura, Y. ^{*4}

*1 School of Engineering and Center for Advanced Metrology, Chubu University, 1200 Matsumoto, Kasugai, Aichi, 487-8501, Japan.

e-mail:txh@isc.chubu.ac.jp

*2 Center for Advanced Metrology, Chubu University, 1200 Matsumoto, Kasugai, Aichi, 487-8501, Japan.

*3 Noritake Company Limited, 3-1-36, noritake, nishi-ku, Nagoya, Aichi, 451-8501, Japan.

*4 Division of Mechanical and Space Engineering, Graduate School of Engineering, Hokkaido University, N 13 W 8, Kita-ku, Sapporo 060-8628, Japan.

Abstract : The present study has been carried out with an aim to develop a measurement technique of two-dimensional gas temperature profiles based on two color fluorescence induced by one color laser. The laser sheet of the fourth harmonics (266 nm) from a Nd:YAG laser has been produced to induce fluorescence from fluorescent materials doped in a nitrogen gas flow. The LIF spectra of seven fluorescent materials, namely acetone, methylethylketone, acetaldehyde ethylbenzene, anisole, aniline, and naphthalene, have been measured to select the best prospective pair of fluorescent materials for this technique. Ethylbenzene and naphthalene show relatively high LIF intensities and are less overlapped in their LIF spectra with each other. 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, 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 way to find out an optimum pair of wavelengths to obtain a good temperature sensitivity in those LIF spectra is presented.

Keywords : temperature measurement, laser induced fluorescence, fluorescent material, temperature sensitivity, laser sheet.

1. Introduction

The laser-induced fluorescence (LIF) technique has been widely applied for diagnostics on reactive fluid systems. Fluorescent materials seeded in the flow were excited by a laser to attain qualitative or quantitative visualizations. As for combustion study, it was applied for visualizations of flow-fields, temperature-fields, pressure-fields, flame-fronts, fuel mixing and fuel evaporation processes, etc. The most common flow-field tracers are acetaldehyde, acetone, biacetyl, and TMPD (Ossler and Aldén, 1997). Acetone is also one of preferable tracers for visualization of fuel mixing and evaporation processes because its pyrolysis does not become noticeable at a temperature even as high as 900 K if a residence time is not very long (Thurber et al. 1998). However, its evaporation characteristics were too dissimilar with those of isooctane which is used as a standard liquid fuel in combustion engine experiments. Since evaporation characteristics of 3-pentanone are very similar with those of isooctane, 3-pentanone was considered a better tracer than acetone to be doped in a liquid fuel. As tracers for the visualizations of fuel mixing and evaporation processes, an exciplex of TMPD and naphthalene was also a prospective one because separate measurements of LIF from injected liquid fuel and its vapor may be possible (Melton, 1983).

As for visualization of temperature-field, 3-pentanone was also considered better tracers than acetone (Grossmann et al. 1996). The fluorescence intensity from hydrocarbon, specifically acetone, was modeled by Thurber et al. (1998). For visualization of temperature-field in combustion flows, nitric oxide has been one of popular tracers (e.g. Seitzman et al. 1985). For visualization of temperature-field in a rarefied gas flow, iodine was used as a tracer (Ni-Imi et al., 1990).

The present study aims at developing a measurement technique of two-dimensional gas temperature profiles based on two color fluorescences from two materials induced by one color laser. Although the technique with a similar concept was applied several times for liquid (Sakakibara et al., 1997, Funatani et al., 2004), its application for gas has not established yet. The present technique would be limited to an isobaric condition because the pressure dependences of two fluorescent materials 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 in an automobile engine room or around a heat transfer devices, such as a room air-conditioner.

Making quantitative visualization of temperature-field using LIF techniques has encountered significant difficulties. One of the reasons for these difficulties comes from the fact that the intensity of laser induced fluorescence depends not only on the temperature but also the number density and optical characteristics of a fluorescent material, and the laser intensity. The laser intensity distribution on a laser sheet is usually not uniform, due to absorption or dispersion by gas molecules in addition to intensity distribution of laser sheet itself, which leads to a measurement error of gas temperature. One of the primary motivations of the present study is to try to reduce this error. In the present measurement technique, 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 materials. Thus, one of the merits of this technique is that the influence of laser intensity distribution on a sheet is canceled out since the laser is emitted from a single laser system.

In choosing excitation wavelength, special attention was paid on the possibility to form a wide laser sheet having high intensity without excessive cost. For this purpose, we focused on 266 nm which is emitted as the fourth harmonics of a Nd:YAG laser. At the first stage of the development, it is necessary to select a pair of fluorescent materials and fluorescent wavelengths. Seven fluorescent materials were selected on the premise that they are excited at 266 nm. LIF spectra of the seven fluorescent materials in nitrogen gas flow were then measured. Temperature dependences of LIF spectra were measured to find out, first an optimum pair of fluorescent materials and then an optimum pair of fluorescent wavelengths for the selected materials.

2. Experimental setup and method

Figure 1 shows an optical layout of an excitation light source. A laser pulse of the second harmonics (532nm) 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). 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 material by bubbling in a doping chamber, as shown in Fig. 2(b), or doped with a solid fluorescent material by going through a solid fluorescent material 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 materials 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 materials, 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 materials. 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.

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 ϕ 1.6 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 materials 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 materials shown in Fig. 3(a) are broadly distributed from 300 nm to 650 nm with relatively low intensities. The LIF spectra of fluorescent materials 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 materials. However, the range of its spectrum is overlapped with other two fluorescent materials shown in Fig. 3(b). On the other hand, the distributions of ethylbenzene and naphthalene are less overlapped with each other and have relatively high LIF intensities. Thus, we selected ethylbenzene and naphthalene as one of prospective pair of fluorescent materials from these seven fluorescent materials 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 materials are sufficient for the purpose of our 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 only a few nm to longer wavelengths. The ranges of LIF spectra also did not show any obvious shifts.

The LIF intensities of both materials 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.

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 materials, and the fluorescence quantum yield. Number densities of fluorescent materials decrease as temperature is increased due to thermal expansion. Because number densities of both materials 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 materials and the fluorescence quantum yield.

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 materials, 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 notation I is the LIF intensity, and (II) the LIF intensity which decides the signal-to-noise ratio of the measurement.

Regarding the first point (I) above, we first discuss the case when there is a single fluorescent material. 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).

Then, we discuss the case when the two fluorescent materials are mixed into a gas flow. We define a function R as $R(\lambda_s, \lambda_l, T) \equiv I(\lambda_s, T)/I(\lambda_l, T)$. The ratio of peak intensities of LIF spectra of both materials depends on the material 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 materials to

have the same peak intensities, $R(\lambda_s, \lambda_l, T)$ becomes

$$R(\lambda_s, \lambda_l, T) = \frac{I_e(\lambda_s, T)/I_{e0} + I_n(\lambda_s, T)/I_{n0}}{I_e(\lambda_l, T)/I_{e0} + I_n(\lambda_l, T)/I_{n0}}, \quad (1)$$

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 $R(\lambda_s, \lambda_l, T)$ gives the temperature sensitivity of the present technique between T_1 and T_2 , namely

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

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

Figure 7 shows equi-contour plots of $\Delta R^*(\lambda_s, \lambda_l, 295 \text{ K}, 374 \text{ K})$, namely the rate-of-change of $R(\lambda_s, \lambda_l, T)$ at the temperatures of $T_1 = 295 \text{ K}$ against $T_2 = 374 \text{ K}$, at various values of λ_s and λ_l . From this figure, the optimum wavelengths λ_s and λ_l were found to be 267 nm and 400 nm, respectively, where $\Delta R^*(\lambda_s, \lambda_l, 295 \text{ K}, 374 \text{ K})$ 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 SN 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 SN 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-2). Thus, this wavelength is to be determined in practice as long a wavelength as possible as far as an S/N ratio of the measurement allows.

Once the optimum pair of wavelengths has been determined, the next point is to obtain a temperature-intensity ratio 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 assures 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 from its value at 295 K. This means that the mean decrease of LIF intensity per unit temperature increment is about 0.9%.

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 material 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 materials.

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 selection 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 harmonics (266 nm) from a Nd:YAG laser was applied to induce fluorescence from fluorescent materials doped in a nitrogen gas flow. LIF spectra of seven fluorescent materials: namely, acetone, methylethylketone, acetaldehyde ethylbenzene, anisole, aniline, and naphthalene, were measured to select the best prospective pair of fluorescent materials for this technique. Ethylbenzene and naphthalene show relatively high LIF intensities and are less overlapped with each other. The spectral measurements at the gas temperatures of 290 K to 387 K revealed that this pair had a preferable characteristic, namely ethylbenzene had a higher temperature dependence while naphthalene had a lower temperature dependence. This characteristic leads to good temperature sensitivity. Temperature is, in the proposed technique, to be determined by the ratio of LIF intensity of mixture at two wavelengths. By selecting one of wavelengths in the range where ethylbenzene is dominant and another where naphthalene is dominant, good temperature sensitivity was found to be attained.

These results indicate that ethylbenzene and naphthalene is a good pair of fluorescent materials. 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 exist around the shorter edge of ethylbenzene spectra and on longer edge of naphthalene spectra. These results show the feasibility of two-dimensional measurement of gas temperature based on two color fluorescence induced by one color laser. The merits of this technique are that the influence of a laser intensity distribution can be cancelled out using a single laser system. It is to be noted that this technique would be limited to the isobaric condition and to relatively low temperatures because those LIF intensities and measurement accuracies decrease with increasing temperature.

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Author Profile

Taro Hirasawa: He received his B. Eng., M.Eng. and D. Eng. Degrees, all from Mechanical Engineering Department of Keio University in 1995, 1997 and 2000, respectively. He first worked at Case Western Reserve University as research associate in 2000-2002. He is currently assistant professor at Department of Mechanical Engineering, Chubu University since 2002. His research interests are fundamental characteristics of premixed flame, micro combustor, and laser diagnostics.



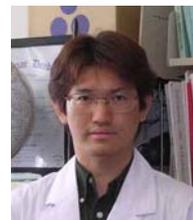
Takahiro Kaneba: He received B. Sc. in Department Physics in 2000 from Ehime University and M. Eng. in Energy Engineering and Science in 2002 from Nagoya University and D. Ac. (Doctor of Academic) in Department of Fusion Science in 2005 from Graduate University for Advanced Studies. He worked in Center for Advanced Metrology, Chubu University as post doctoral fellow. His research interests are gas temperature measurement by LIF.



Yuichi Kamata: He received his Ph.D. in Physics in 1995 from Nagoya University. He worked in Research Center for Advanced Energy Conversion, Nagoya University as a lecturer in 1997 - 2000. He works in Noritake Company Limited as a technical adviser since 2001. His research interests are X-ray astronomy in Astrophysics, Atmospheric and Environmental Science and Thermal Visualization.



Katsunori Muraoka: He received his B. Eng., M.Eng. and D. Eng. Degrees, all from Mechanical Engineering Department of Kyushu University in 1963, 1965 and 1970, respectively. He first worked at Research Institute for Applied Mechanics, and then at Interdisciplinary Graduate School of Engineering Sciences of the same University until his retirement in 2004. He moved to the current position in April 2004. His research interests are plasma physics and gas discharge phenomena, in particular developments of laser-aided diagnostics of plasmas and gases. In this field, he published a book "Laser-aided diagnostics of plasmas and gases" (Institute of Physics Publishing, U. K., 2001).



Yuji Nakamura: He received his M.Sc. (Eng) degree in Mechanical Engineering in 1995 from Nagoya University and his Ph.D. in 2000 from same university. He worked in University of Kentucky as visiting research assistant in 1996-1997. He joined in Department of Mechanical Engineering, Nagoya University as research associate and became associate professor at Center for Integrated Research and Engineering and EcoTopia Science Institute, Nagoya University. He also worked at National Institute of Standards and Technology (NIST) in 2000-2001 as visiting professor. He is currently associate professor at Division of Mechanical and Space Engineering, Hokkaido University since 2005. His research interests are Combustion, Fire Science, and Safety Engineering.

Figure Captions

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.

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

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.

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.

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

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.

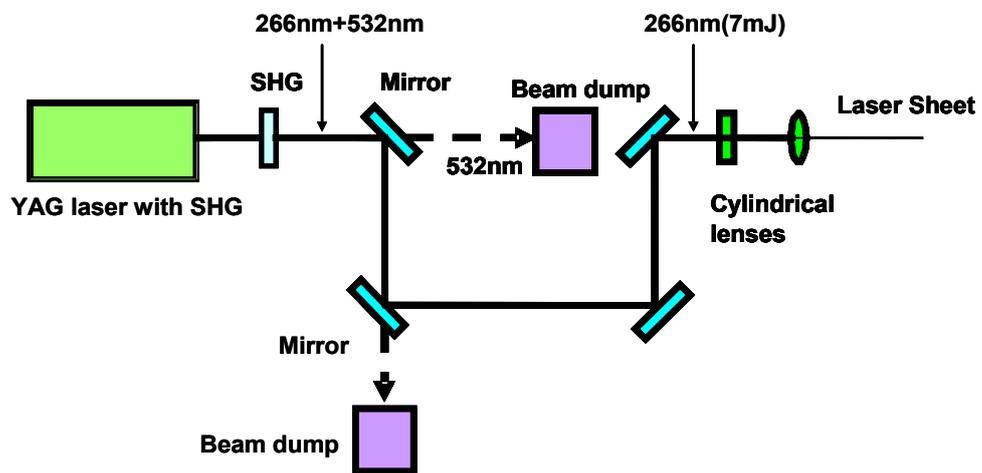


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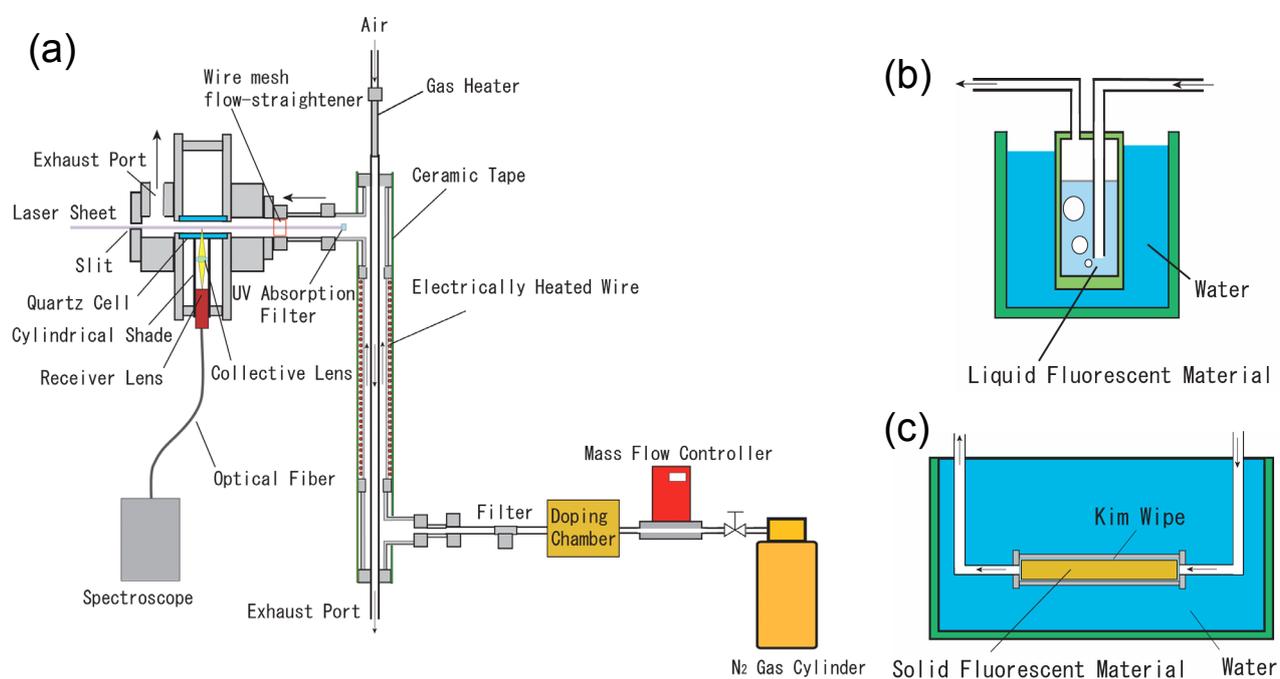


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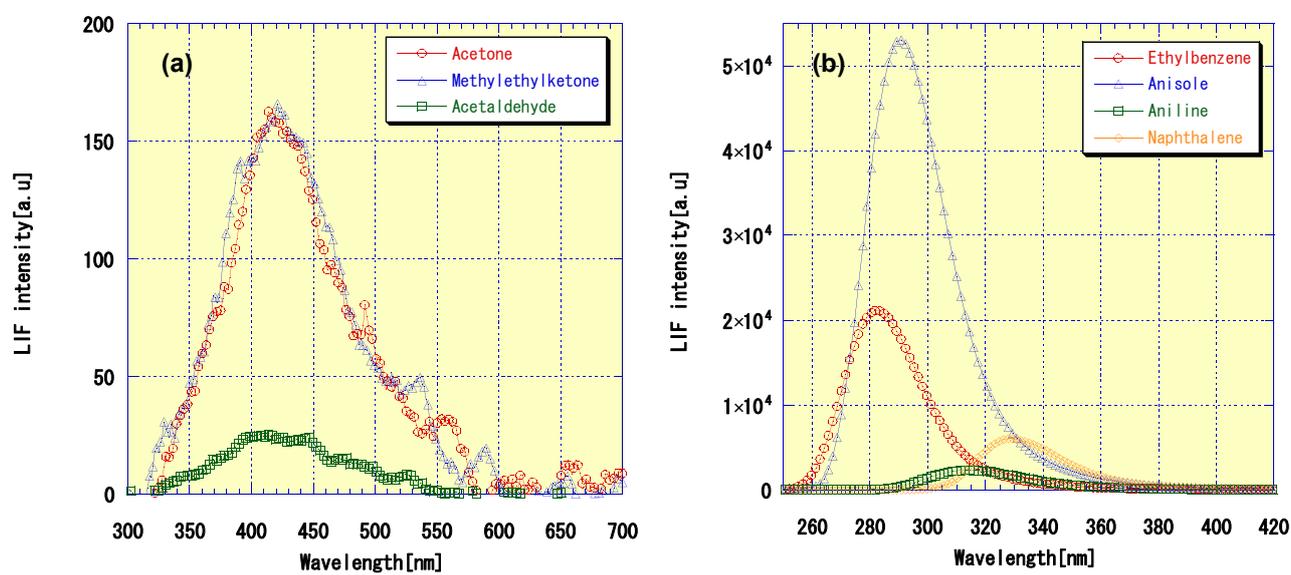


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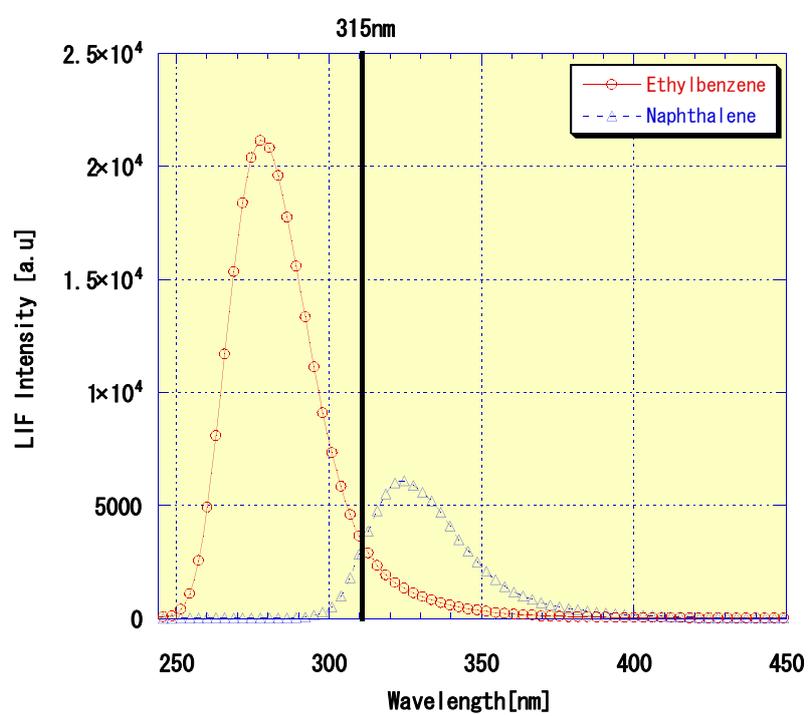


Fig. 4. LIF spectra of ethylbenzene and naphthalene.

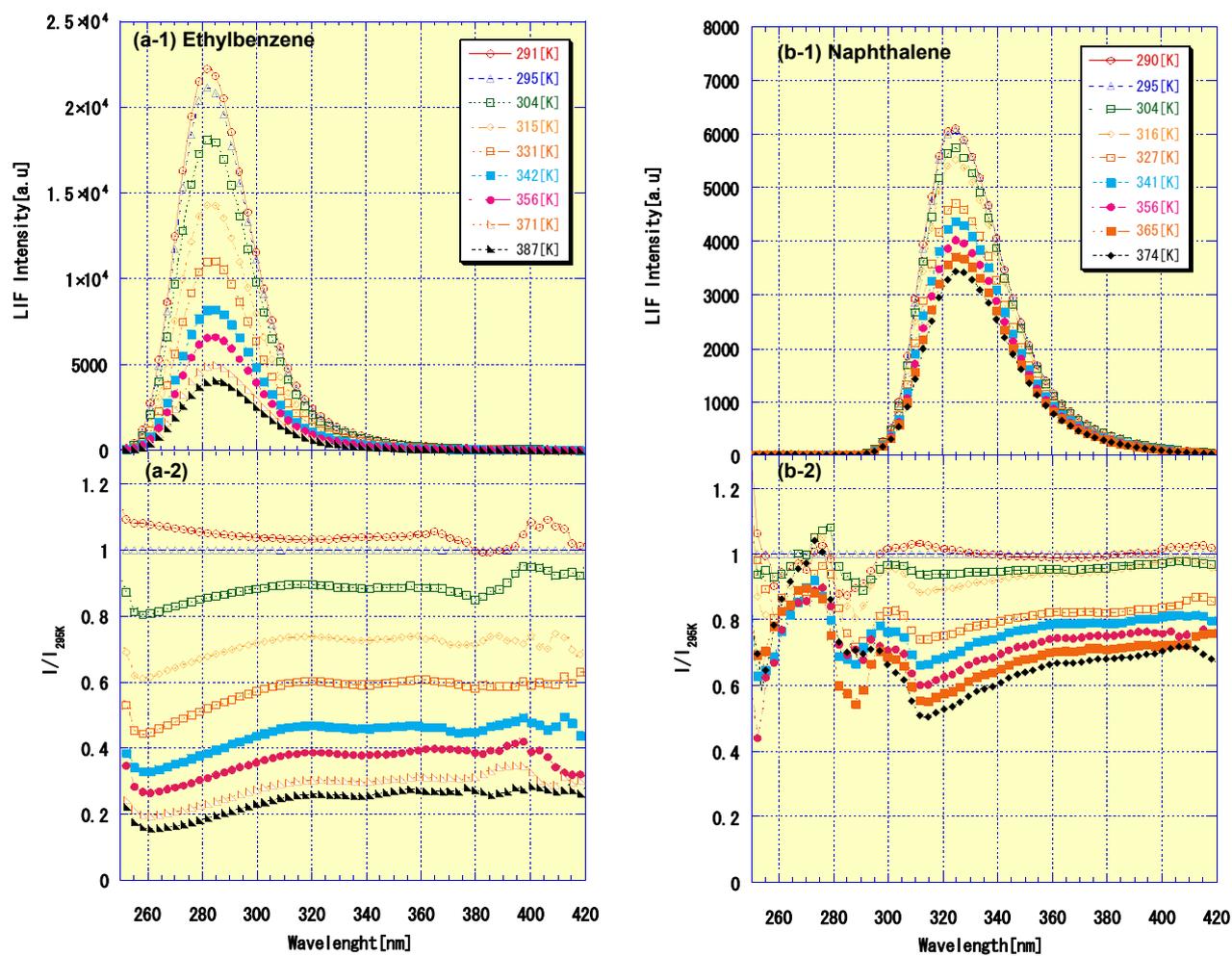


Fig. 5. LIF Spectra at various temperature: (a-1) and (a-2) are for ethylbenzene; (b-1) and (b-2) are for naphthalene; (a-2) and (b-2) are normalized forms relative to the LIF spectra at 295 K.

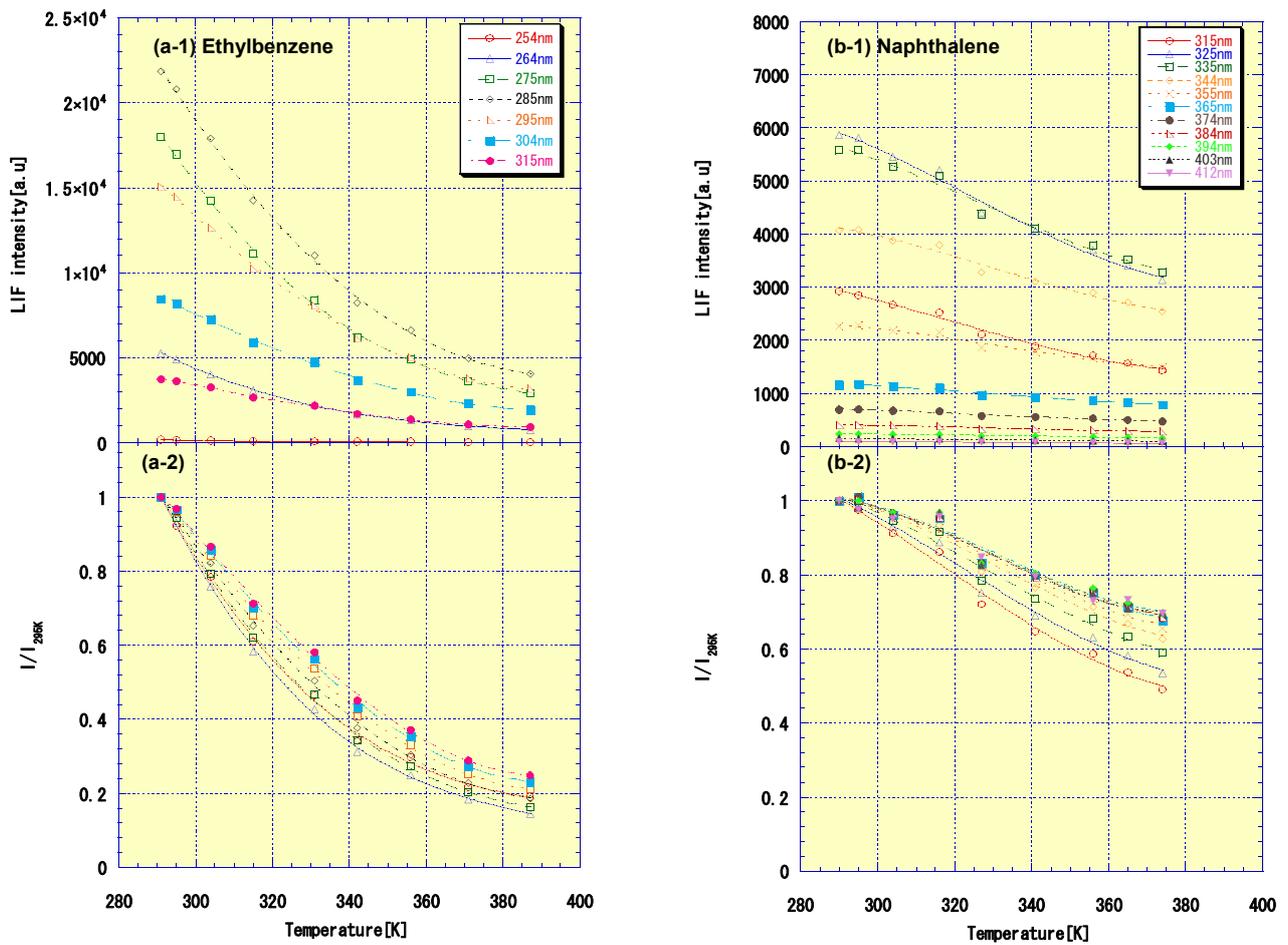


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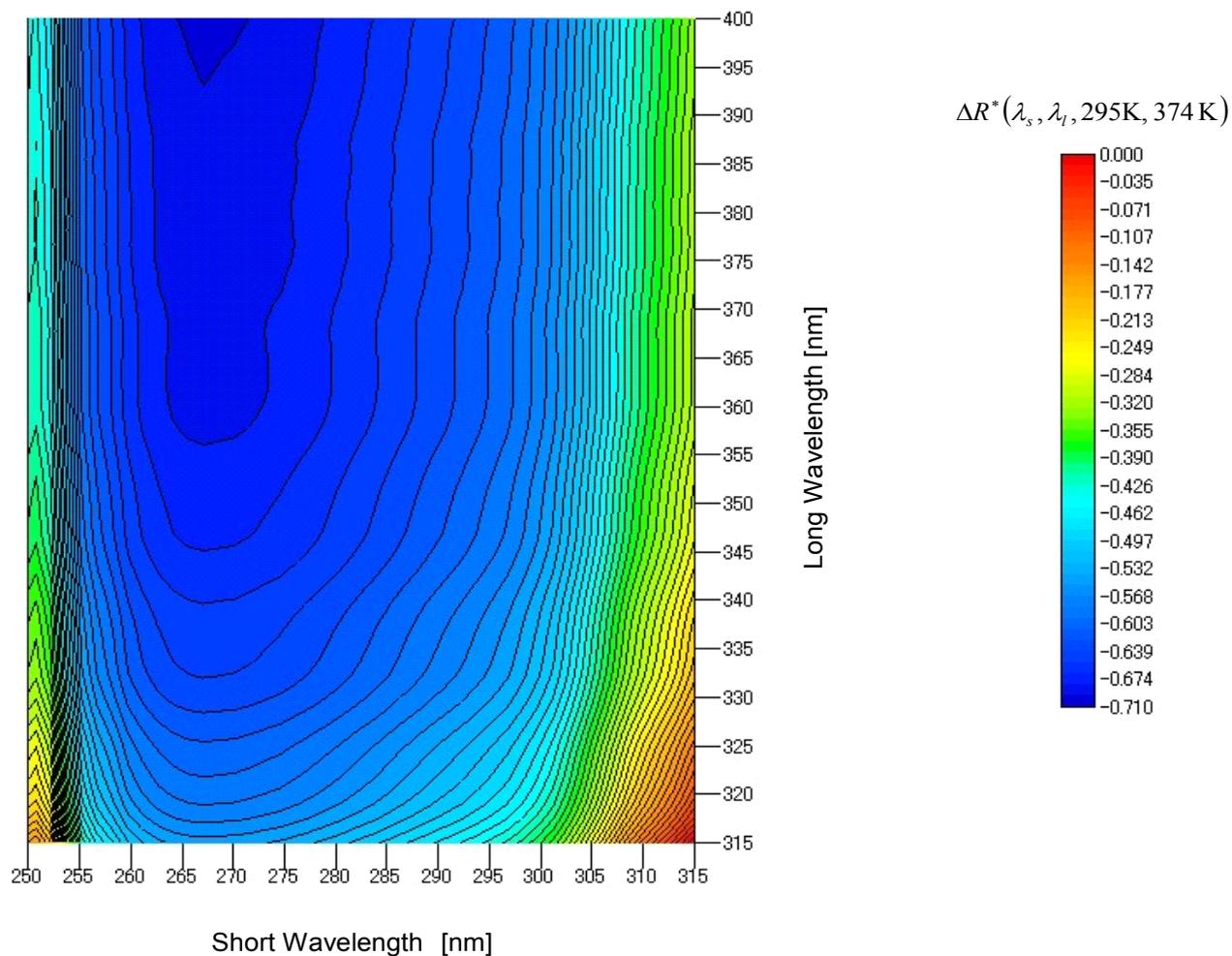


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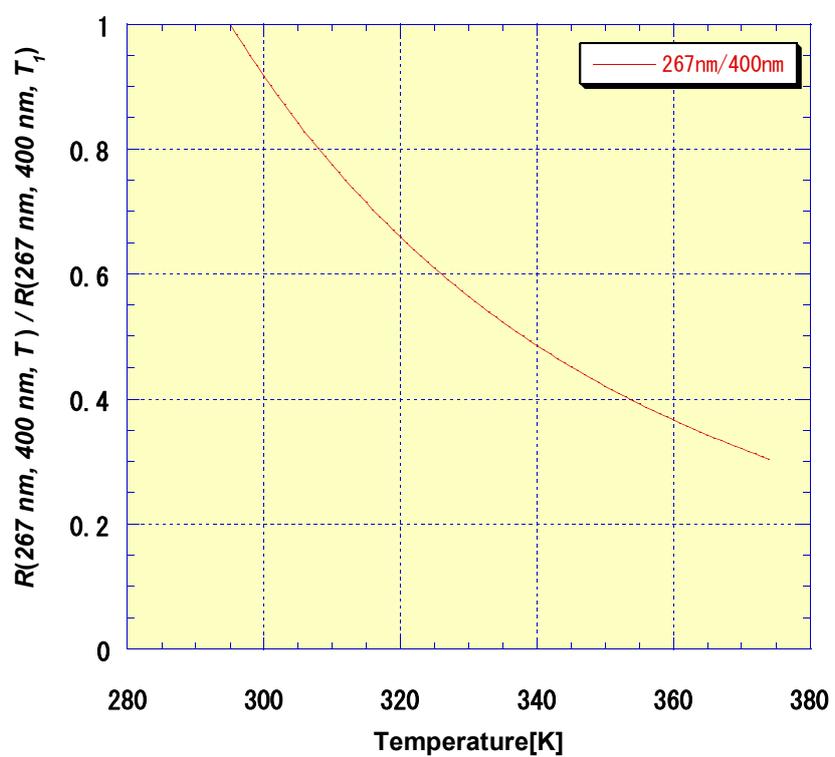


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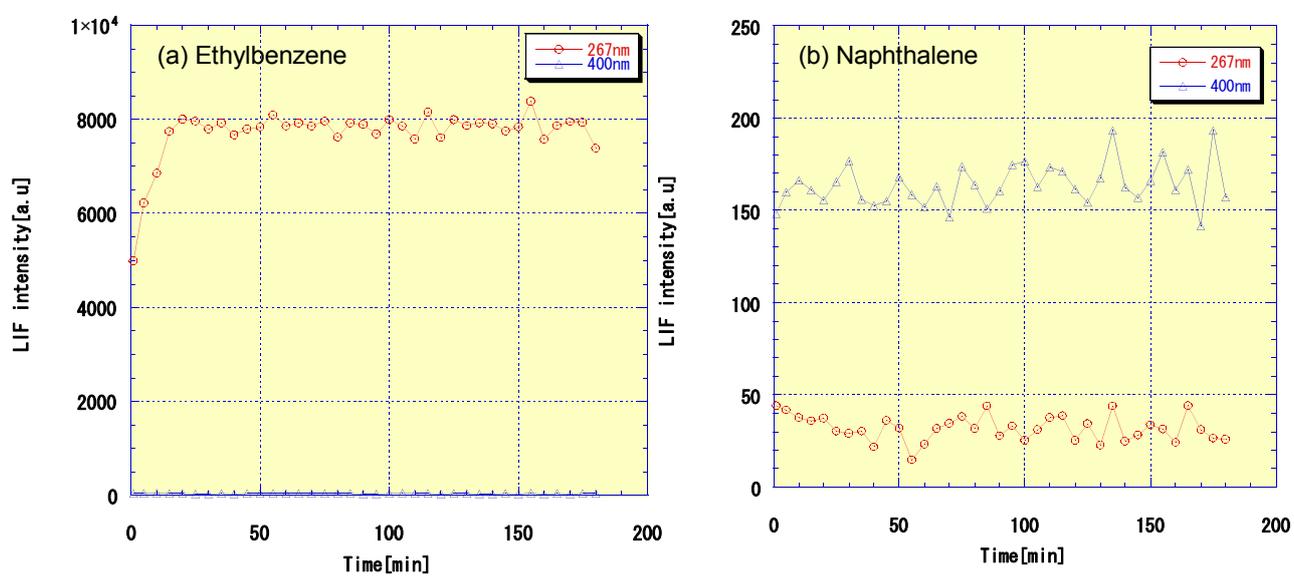


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