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2 **Quantitative measurement of temperature in oxygen enriched**  
3 **CH<sub>4</sub>/O<sub>2</sub>/N<sub>2</sub> premixed flames using Laser Induced Thermal Grating**  
4 **Spectroscopy (LITGS) up to 1.0 MPa**  
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6  
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22 **Colloquium**

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Abstract	186	
Main text	3812	—
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Figure	1 (348), 2 (235), 3 (117), 4 (305), 5 (475), 6 (130), 7 (138)

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36 No color images to be charged are included.

37 **Abstract**

38 The application of laser diagnostics to high pressure combustion phenomena is particularly challenging,  
39 especially in practical combustors such as rocket motors. In this study, temperature measurements  
40 using Laser Induced Thermal Grating Spectroscopy (LITGS) are demonstrated in oxygen enriched  
41 CH<sub>4</sub>/O<sub>2</sub>/N<sub>2</sub> premixed laminar flames at pressures up to 1.0 MPa. We use a previously developed OH  
42 absorption LITGS technique to determine product gas temperatures from 0.3 to 1.0 MPa, for both high  
43 temperature oxygen-enriched and pure-oxygen flames, for measurements up to 3000 K. Further, we  
44 demonstrate how it is necessary to correct the measurements for the local absorption of laser light to  
45 obtain accurate temperatures, and offer a technique for producing the correction by using different  
46 laser energies. Once the correction is applied, we demonstrate that the measurements at 0.5 MPa are  
47 within 1.6% of the adiabatic non-strained flame temperatures, with a standard deviation of about 160  
48 K, thus offering a competitive method for the challenging conditions at high pressures and  
49 temperatures. The values obtained at derived temperatures at 1.0 MPa were slightly lower than the  
50 adiabatic unstrained flame temperatures, which could possibly be attributed to heat losses.

51

52 **Keywords**

53 LITGS; High temperature; High pressure; Quantitative temperature measurement; Oxygen enriched  
54 flames

55

## 56 **1. Introduction**

57 Many combustors in devices such as reciprocating engines, gas turbine and rocket motors operate  
58 at high pressures in order to increase power density. This is particularly true in the case of rocket  
59 motors, where hydrogen-oxygen combustion at pressures of order 10 MPa [1] is employed and  
60 temperatures are expected to be in excess of 3000 K. Yet these extreme pressures and temperatures  
61 render the measurement of scalar quantities particularly challenging, as optical techniques can be  
62 limited by light absorption and spectral broadening [2].

63 Temperature is a key controlling scalar in most combustion systems, so that there is an extensive  
64 research effort to produce quantitative measurements of flame temperatures. These tools include the  
65 use of thermocouples (e.g. [3]), Rayleigh scattering (e.g. [4]), Coherent anti-Stokes Raman  
66 spectroscopy (CARS) (e.g. [5]), Laser Induced Electrostrictive Gratings (LIEGS) (e.g. [6]), and Laser  
67 Induced Thermal Grating Spectroscopy (LITGS) [7-19]. The high pressures prevalent in gas turbines  
68 and rocket systems create significant challenges for optical diagnostics. Whereas on one hand, the  
69 higher concentrations help produce higher signal from the larger number of molecules per unit volume  
70 interacting with light, countervailing effects can reduce the signal to noise ratio of the techniques,  
71 including: (a) background radiant interference from intense chemiluminescence, which interferes with  
72 spectroscopic diagnostics such as fluorescence or Rayleigh scattering, (b) line broadening from  
73 techniques which rely on a well-defined spectrum, such as Planar Laser Induced Fluorescence (PLIF),  
74 and (c) increased signal trapping and quenching from collisions, particularly in the case of fluorescence.

75 In contrast with these techniques, LITGS offers potential advantages for quantitative  
76 measurements of temperature at high pressure. Unlike many other laser-based techniques, the signal  
77 and signal/noise ratio produced increases with pressure. As detailed further on, this is because the  
78 signal intensity is proportional to the square of the induced refractive index fluctuation [15], which  
79 increases with the density of the mixture. A number of studies using LITGS have been used in non-  
80 reacting [8-13] and reacting [14-19] flows. Although LITGS can only be used in the presence of an  
81 absorbing species, LITGS also offers the advantage of significantly higher signal relative to LIEGS,

82 and less stringent requirements on optical alignment relative to CARS.

83 Despite the growing interest associated with LITGS measurements, only a handful of studies have  
84 used LITGS for measuring product gas temperatures. The reasons are: low concentration of species to  
85 be excited in the product gas especially at low pressure; an immediate decay of signal due to high  
86 thermal diffusivity at low pressures. Absorption by the hydroxyl radical OH around 307 nm was used  
87 in measurements of product temperature over a flat-flame burner stabilized using H<sub>2</sub>/O<sub>2</sub> at atmospheric  
88 pressures [14], and CH<sub>4</sub>/air mixtures at pressures from 1.0 to 4.0 MPa [16]. More recently, Sahlberg  
89 *et al.* [18] obtained LITGS signals over an atmospheric pressure ethylene flame using infrared  
90 absorption around 3.2 μm for both the fuel itself and H<sub>2</sub>O to generate the thermal grating. Since fuel  
91 in unburnt mixture and water vapor in product gas are major components, a stronger LITGS signal  
92 than for OH is expected even at low pressure.

93 A comparison of the expected OH concentrations in the experiments by Latzel *et al.* [16] with  
94 those in oxygen-enriched, high temperature flames suggested that the latter might be as high or higher  
95 than in the original experiments, thus allowing higher quality measurements. Therefore, the purpose  
96 of this study is to demonstrate the capability of LITGS using OH excitation for product gas temperature  
97 measurements in high pressure and high temperature, oxygen-enriched flames.

98

## 99 **2. Temperature measurement using OH-LITGS**

### 100 **2.1. Theory of LITGS**

101 The LITGS technique has been described in detail elsewhere (see [7]), so the following  
102 description is limited to the basic theory. In LITGS, pulsed pump laser beams at a wavelength,  $\lambda$ , cross  
103 at an angle  $2\theta_p$  at a probe volume, generating a grating of spacing  $\Lambda = \lambda/(2\sin\theta_p)$ . The pump  
104 wavelength targets a specific transition for molecular excitation of a particular species of interest, so  
105 that the molecules in the regions of high intensity within the grating absorb energy during the short  
106 (nanoseconds) laser pulse. Part of the energy absorbed is released via collisional quenching in the form  
107 of thermal energy, generating a stationary temperature and density perturbation, in the form of a

108 thermal grating across the interacting region, which decays due to the thermal diffusion over hundreds  
 109 of nanoseconds. The rapid temperature change generates an acoustic wave which propagates locally  
 110 through the thermal grating. The interference of traveling acoustic and stationary density perturbations  
 111 creates a local density oscillation. The latter is probed by using a continuous wave laser beam of a  
 112 different wavelength,  $\lambda_p$ , directed at the Bragg angle,  $\theta_b = \sin^{-1}[\lambda_p/(2\Lambda)]$ . Bragg diffraction off the  
 113 grating generates a coherent signal beam which, when collected, yields the local temporal evolution  
 114 of the overall density oscillation.

115 The frequency of the LITGS signal oscillation,  $f_{osc}$ , is equal to the inverse time required for the  
 116 passage of the sound wave through the grating spacing  $\Lambda$  at a velocity,  $a$ ,

$$117 \quad f_{osc} = \frac{a}{\Lambda}. \quad (1)$$

118 The speed of sound in an ideal gas is given by:

$$119 \quad a = \sqrt{\frac{\gamma R^* T}{W}}, \quad (2)$$

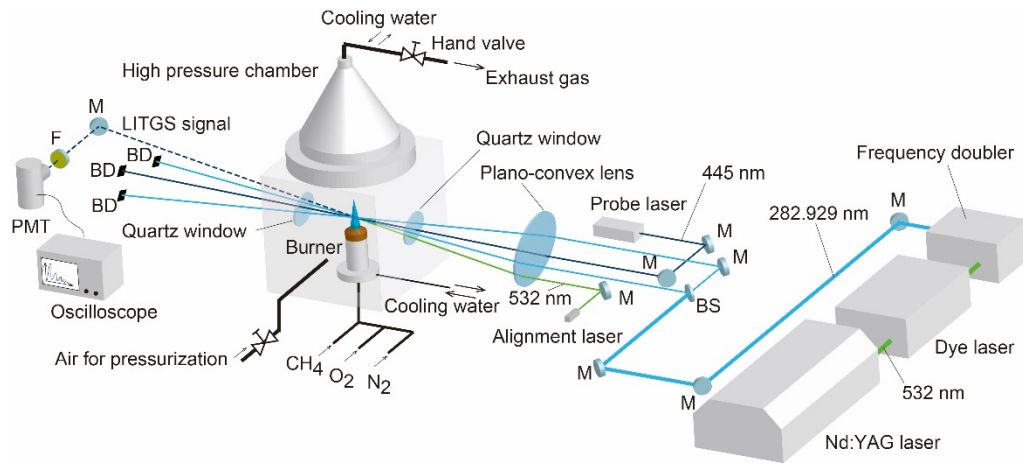
120 where  $\gamma$ ,  $R^*$ ,  $W$  and  $T$  are the specific heat ratio, the universal gas constant, mean molecular weight and  
 121 temperature, respectively. From Eqs. (1) and (2), the temperature within the probe volume can finally  
 122 be determined as:

$$123 \quad T = \frac{W}{\gamma R^*} \Lambda^2 f_{osc}^2. \quad (3)$$

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131 **2.2. Experimental setup and experimental conditions**

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133

134 *Figure 1. Experimental setup and the layout of the optical geometry through the high pressure*  
 135 *chamber for high pressure OH-LITGS experiments. The symbols M, BS, BD, F and PMT represent*  
 136 *mirror, beam splitter, beam dump, interference filter and photo multiplier tube, respectively.*

137

138 Figure 1 shows the experimental setup and the layout of the optical geometry through the high  
 139 pressure combustion chamber. The high pressure combustion experiments were carried out using the  
 140 combustion test facility at the Institute of Fluid Science, Tohoku University. Details of the test facility  
 141 have been described elsewhere [20]. A Nd:YAG laser (Spectra-Physics, GCR-250, 532 nm) pumping  
 142 a frequency doubled dye laser (Lumonics, HD500+HT1000) was employed to obtain a pump beam set  
 143 to a wavelength of 282.929 nm for the  $Q_1(6)$  branch of OH(1,0) band excitation. The pump energy for  
 144 OH excitation was measured before the beam splitter and set to 9 mJ. The pump beam was split using  
 145 a 50:50 beam splitter to separate two laser beams, and the parallel beams are introduced via a plano-  
 146 convex lens (OptoSigma, focal length,  $F=1000$  mm at 546.1 nm), termed as the crossing lens in this  
 147 paper. The distance between the two pump laser beams,  $\delta$ , was set to 10 mm. The two beams are  
 148 crossed at the measurement point with an angle of  $2\theta_p$ . The refractive index inside the chamber and of  
 149 the flame changes slightly with changes in the pressure inside the chamber, and so does the optimum  
 150 alignment. In order to maximize the LITGS signal, the alignment was adjusted before the series of

151 experiments at the same pressure condition.

152 A continuous wave (CW) semiconductor laser, operating at a wavelength of 445 nm (RGB laser  
153 system, NavoPro PB 445-1000 MM) was employed as the probe beam. The power for the probe beam  
154 was set to 810 mW. The beams passing through the high pressure chamber were stopped by beam  
155 dumps. The diffracted signal (LITGS signal) was detected using a photo multiplier tube (PMT,  
156 Hamamatsu Photonics, R928) through an interference filter (OptoSigma, YIF-BA420-460S) that  
157 transmits the beam at 445 nm. The LITGS signal was acquired using an oscilloscope with a sampling  
158 rate of 10 GS/s and 500 MHz bandwidth (Keysight, DSOS054A).

159 The pump beam diameter at the crossing lens was measured at the crossing point and found to be  
160 slightly elliptical, with major and minor axes around 4.7 mm and 3.4 mm, respectively. By assuming  
161 a Gaussian profile of pump beams, the lengths of the measurement volume in the forward beam axis  
162 direction and two perpendicular directions were about  $19.0 \times 0.1 \times 0.1$  mm. The relatively long focal  
163 length of the crossing lens employed in this study yields a relatively long measurement volume.  
164 However, the actual measurement point volume is the intersected volume of the two pump beams and  
165 the probe beam, and is thus smaller than that implied by the values above. In this study, measurements  
166 were performed at the center of the burner, where the temperature gradients within the measurement  
167 volume are expected to be small according to the flame observation by OH-PLIF that will be shown  
168 in Section 3.1. Smaller probe volumes could of course be realized by increasing the crossing angle,  $\theta_p$ ,  
169 but require a different optical arrangement with a larger crossing lens with a shorter focal length.

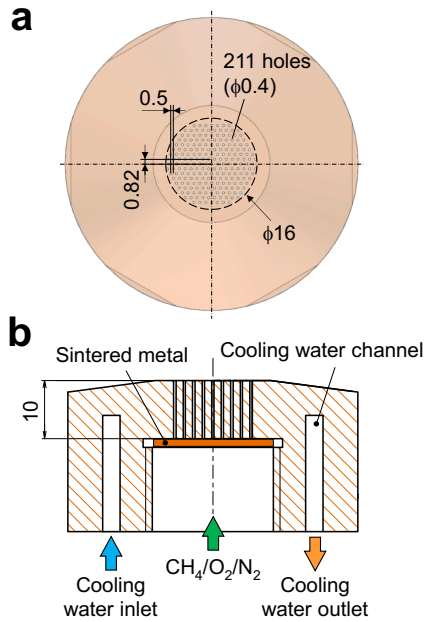


Figure 2. Schematic figures of the modified burner nozzle for the calibration burner developed by Takeuchi *et al.* [21]: (a) Top-view; (b) Cross-sectional-view.

Oxygen enriched mixtures of CH<sub>4</sub>, O<sub>2</sub> and N<sub>2</sub> were considered in this study. The oxygen ratio in the (O<sub>2</sub>+N<sub>2</sub>) mixture is denoted as,  $\beta = Q_{O_2} / (Q_{N_2} + Q_{O_2})$ , where  $Q_x$  is the volumetric flow rate of species  $x$ . The calibration burner available at high pressure developed by Takeuchi *et al.* [21] was employed in this study. A modified burner nozzle which is schematically shown in Fig. 2 was mounted in order to prevent from flashback of oxygen enriched flame. The burner nozzle was made of oxygen-free copper and was water cooled. The nozzle has 211 holes, each with a diameter of 0.4 mm, drilled within a 16 mm circular exit. The measurement points were made at a centerline position located 15 mm downstream of the burner surface. Experiments were performed at pressures,  $P$ , of 0.3, 0.5 and 1.0 MPa, and the reactant temperature was 298 K. The equivalence ratio was varied from 0.6 to 1.2 for CH<sub>4</sub>/O<sub>2</sub>/N<sub>2</sub> flames and 0.3 to 0.6 for CH<sub>4</sub>/O<sub>2</sub> flames.

### 3. Results and discussions

#### 3.1. Flame characteristics stabilized on the calibration burner



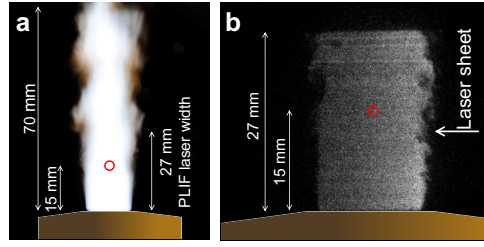


Figure 3. (a) Flame image of direct photo and (b) instantaneous OH-PLIF image at  $P = 1.0$  MPa for  $\phi = 1.0$  and  $\beta = 0.6$ .

Figure 3 shows the direct photo and OH-PLIF image of a flame stabilized on the calibration burner at  $P = 1.0$  MPa for  $\phi = 1.0$  and  $\beta = 0.6$ . OH-PLIF images were collected at 263.580 nm excitation for OH(2,0) band excitation [2, 21] in order to reduce the influence of laser attenuation. The laser energy for OH-PLIF was 14 mJ. The circle in Fig. 3 represents the center of the measurement point of LITGS. The broadband white emission in the downstream region of the oxygen-enriched flames could be observed, whereas the interfaces of the hole-stabilized flames on the burner surface were too small to be resolved. The Reynolds numbers based on the hole diameter and mean velocity were about 3000 at 1.0 MPa, and thus somewhat higher than the critical transitional Reynolds number, making the flow weakly turbulent. However, as shown in Fig. 3b, the burned gas downstream the burner was uniform. At other pressure conditions, the Reynolds numbers were smaller than 2300.

Numerical simulations were performed using CHEMKIN-PRO [22] with GRI-Mech 3.0 [23] to determine expected species molar fractions and temperatures. The experimentally measured heat loss to the burner was estimated from the flow rate of water and the inlet and outlet water temperatures. The estimated ratio of heat loss to the burner as a fraction of the heat release was varied from 1.5-9.9%. Adiabatic non-stretched calculations were used as a reference for the numerical simulation, yet the heat loss ratio suggests the calculated values should be considered an upper bound. The maximum expected molar fraction of OH at a height above burner 15 mm downstream of the point of peak heat release rate reaches about 5% for  $\beta = 0.6$  and 10% for  $\beta = 1.0$ .

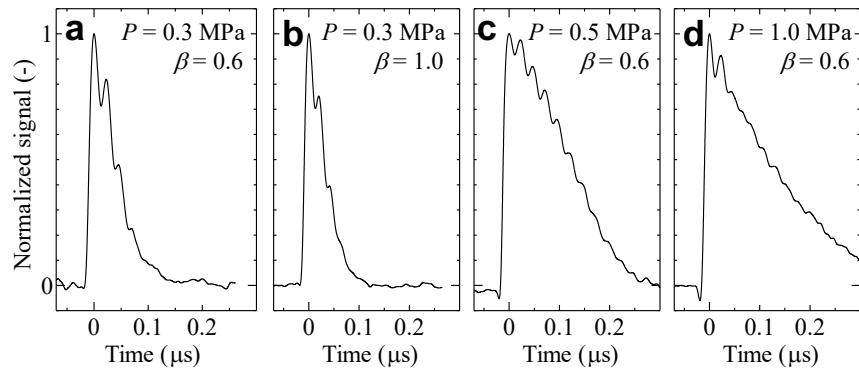
The measurements of OH-PLIF confirm that OH fluorescence levels are uniform around the

210 measurement point [21], although the fluorescence levels decrease strongly from right to left due to  
211 laser beam intensity decrease by absorption because of the high OH concentrations at 1.0 MPa.

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### 213 3.2. Derived temperature from LITGS signal

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215

216 *Figure 4. Representative single shot LITGS signal for the equivalence ratio of  $\phi = 0.6$ : (a)  $P$*   
217 *= 0.3 MPa,  $\beta = 0.6$ ; (b)  $P = 0.3$  MPa,  $\beta = 1.0$ ; (c)  $P = 0.5$  MPa,  $\beta = 0.6$ ; (d)  $P = 1.0$  MPa,  $\beta = 0.6$ .*

218

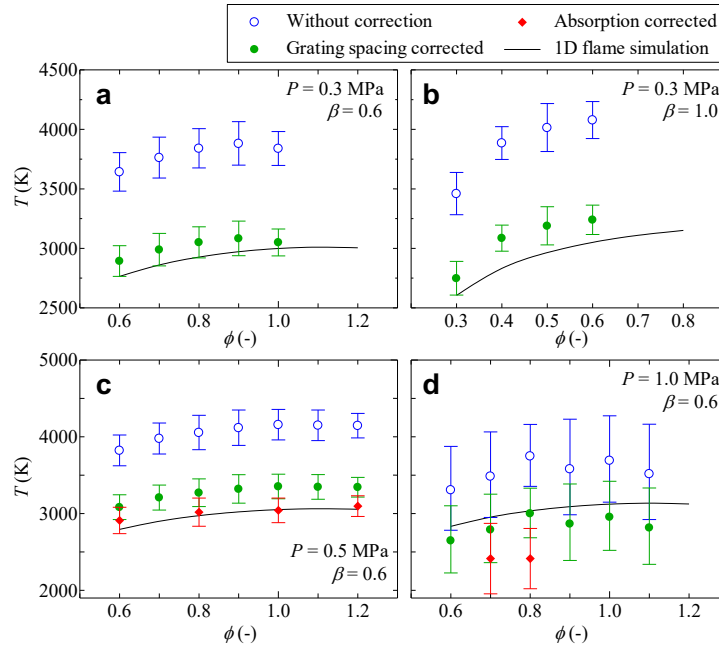
219 Figure 4 shows single shot LITGS signals. Here, the zero time origin is defined as the instant of  
220 the first local peak of the LITGS signal. As mentioned in Section 2.2, the alignment was slightly  
221 adjusted before the series of constant pressure conditions, so the LITGS signals could not be exactly  
222 directly compared under various pressure conditions. Therefore, the representative signal normalized  
223 by the peak value in each case is shown in the figure. The increase in pressure (and density) leads to a  
224 slower decay in the LITGS signal, as the thermal diffusivity decreases. Only a few clear peaks were  
225 observed (Figs. 4a and 4b) because of significantly fast decay of LITGS signal at 0.3 MPa. The number  
226 of local peaks and troughs observed increased at 0.5 MPa (Fig. 4c). However, the contrast between  
227 peaks and troughs was poorer for 1.0 MPa than 0.5 MPa. Although this may require further  
228 investigation, the high concentration of water vapor inside the high pressure conditions led to  
229 condensation on the windows and poorer signal quality (Fig. 4d).

230 The oscillation frequency,  $f_{osc}$ , was determined as the averaged value of the frequency obtained

231 as the inverse of the time between the first two local peaks and troughs rather than a Fourier transform.  
232 In some borderline cases, LITGS signals had small and fast decaying oscillation, as shown in Fig. 4b.  
233 A total of 512 traces were acquired for all experimental conditions but only the signals that clearly  
234 contained two local peaks and troughs were used for the analysis, resulting in a yield of 36 to 511  
235 traces used to determine the oscillation frequency, depending on the experimental condition. The mean  
236 oscillation frequency and standard deviation were determined from the collected signals. This value  
237 was squared, and the temperature evaluated using Eq. (3). Initially, the grating spacing  $\Lambda$  was evaluated  
238 based on the distance of two pump beams and the nominal value of focal length of the crossing lens  
239 (1000 mm).

240 Equation (3) shows that the estimate of temperature depends on the values of  $W/\gamma$ , which is a  
241 function of composition and temperature. In this study,  $W/\gamma$  values were calculated based on the  
242 temperature and mixture composition calculated using the CHEMKIN simulation at 15 mm  
243 downstream from the position of maximum heat release rate in unstrained, adiabatic free flame  
244 simulations. As discussed in Section 3.1, heat loss is not entirely negligible, yet the sensitivity of the  
245 values of  $W/\gamma$  to heat loss is small. A decrease in the assumed temperature by a maximum of 10.2%,  
246 corresponding to the maximum heat loss, leads to a change in  $W/\gamma$  of 6.7% at the condition of  $P = 0.5$   
247 MPa,  $\beta = 0.6$  and  $\phi = 1.0$ . A change in the equivalence ratio of reactants by 1% leads to a change in  $\gamma$   
248 and  $W$  of less than 0.5%.

249



250  
 251 *Figure 5. Derived temperature from the LITGS signals with and without corrections (see*  
 252 *text): (a)  $P = 0.3$  MPa,  $\beta = 0.6$ ; (b)  $P = 0.3$  MPa,  $\beta = 1.0$ ; (c)  $P = 0.5$  MPa,  $\beta = 0.6$ ; (d)  $P = 1.0$*   
 253 *MPa,  $\beta = 0.6$ . Solid lines are obtained from unstrained flame simulations.*

254  
 255 The open circle symbols in Fig. 5 shows the derived LITGS temperature as a function of  
 256 equivalence ratio and oxygen ratio. As mentioned above, the signal to noise ratio at 1.0 MPa was  
 257 relatively small compared to other pressure conditions. Therefore, the error bars of the derived  
 258 temperature were relatively wider and larger than the results by Latzel *et al.* [16] because of the  
 259 influence on high concentration of water vapor inside the chamber and the small amount of water  
 260 vapor condensed on the windows. Clearly, the measured LITGS temperature were 500 - 1000 K higher  
 261 than the temperature calculated from CHEMKIN simulations. Whereas lower temperatures might have  
 262 been justified from heat loss calculations, higher temperatures can hardly be justified, and the reason  
 263 for the discrepancy must lie elsewhere.

## 264 265 4. Temperature correction

### 266 4.1. Grating spacing correction

267 The temperatures derived from Eq. (3), depend on the accuracy of the determination of the

268 properties of the mixture ( $\gamma$  and  $W$ ), grating spacing,  $\Lambda$ , and oscillation frequency,  $f_{osc}$ . The value of  
269 grating spacing is particularly important because the derived temperature is proportional to the square  
270 of grating spacing as shown in Eq. (3).

271 In the original analysis, the grating spacing,  $\Lambda$ , was determined by the geometric incident angle,  
272 which is a function of the pump beam separation and the focal length of the crossing lens. The focal  
273 length of the crossing lens used in this study was specified at a wavelength of 546.1 nm, and the actual  
274 focal length was therefore about 10% shorter than the nominal value because of chromatic aberration,  
275 resulting in an originally estimated grating spacing of 28.3  $\mu\text{m}$ . In order to obtain a more precise  
276 estimate, an atmospheric pressure measurement of temperature using acetone vapor/air non-reacting  
277 mixture was done for calibration purposes [19, 24]. Acetone vapor was generated by a gas bubbler,  
278 and diluted using dry air down to a value known not to affect the local temperature measurements by  
279 local absorption, as discussed in Section 4.2. The local temperature at the outlet of the nozzle for  
280 acetone vapor/air mixture was measured by a thermocouple for calibration. As shown in Section 4.2,  
281 the derived temperature from acetone vapor/air mixture was influenced by the laser energy and acetone  
282 concentration. The acetone mole fraction varied from 0.005 to 0.012, but the corresponding change in  
283 derived temperature was less than 4 K. Therefore, the influence on laser power and acetone  
284 concentration could be considered small, and the grating spacing calibration was performed for the  
285 acetone mole fraction of 0.012, as the acquired LITGS signal was best at this acetone mole fraction.  
286 By using the measured temperature at the LITGS measurement point and acquired oscillation  
287 frequency, the actual grating spacing was determined from Eq. (3) as  $25.3 \pm 1 \mu\text{m}$  depending on the  
288 LITGS alignment. The focal length calculated from the measured grating spacing and the crossing  
289 angle calculated from the measured value were  $894 \pm 4 \text{ mm}$  and  $0.64 \pm 0.02^\circ$ , respectively.

290 The closed circle symbols in Fig. 5 represent the LITGS temperature calculated using the value  
291 of the calibrated grating spacing above rather than the originally assumed value. The revised  
292 temperatures are closer to the simulated values, especially at  $P = 0.3 \text{ MPa}$ . In the case at  $P = 1.0 \text{ MPa}$ ,  
293 however, the derived temperatures were lower than that of the value of CHEMKIN simulations.

294

## 295 **4.2. Correction for laser light heating**

296 There is a tradeoff in obtaining robust LITGS signals by using high pulse energy, and the  
297 generation of a local disturbance to the local temperature and density because LITGS is an absorption  
298 based technique. A previous study [12] showed that the derived temperature from LITGS in acetone  
299 or toluene increased linearly with an increase in concentration of absorbing media times laser energy.  
300 Therefore, the influence of absorption should be considered when the incident laser energy and/or  
301 concentration of absorbing media are sufficiently high. The total energy absorbed by the gas per unit  
302 area,  $\Delta E$  within a probe volume of length  $L$ , can be estimated by the Beer-Lambert Law as [24],

$$303 \quad \Delta E = E_0 \{1 - \exp(-\sigma N_A n_{OH} L)\} \approx E_0 \sigma N_A n_{OH} L, \quad (4)$$

304 where  $\sigma$ ,  $N_A$ ,  $n_{OH}$  and  $E_0$  are the absorption cross section, Avogadro's constant, the molar concentration  
305 of OH and the total energy deposited, respectively. In the case where absorption is low, the exponential  
306 term can be expanded, resulting in a linear behavior with the concentration of the absorbing species  
307 and the initial energy. Considering that the probe volume heat capacity is  $mc_p$  ( $m$ : mass,  $c_p$ : mixture  
308 averaged specific heat at constant pressure), the relative temperature rise by absorption of the incident  
309 laser,  $\Delta T$ , can be formulated as

$$310 \quad \frac{\Delta T}{T_0} \propto \frac{\Delta E}{mc_p T_0} = \frac{E_0}{c_p T_0} \cdot \frac{\sigma N_A L}{VW} \cdot \frac{n_{OH}}{n} \propto X_{OH} E_0, \quad (5)$$

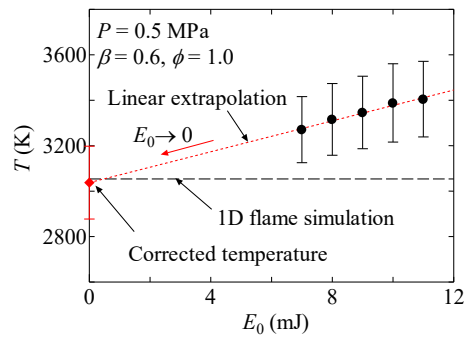
311 where  $V$ ,  $n$ ,  $X_{OH}$  are the probe volume, the molar density of mixture and the mole fraction of OH,  
312 respectively.

313 We can therefore obtain the original temperature with no laser absorption by obtaining  
314 measurements with different energies, and extrapolating the obtained LITGS frequency to the limit of  
315  $E_0 \rightarrow 0$ .

316 Figure 6 shows the relationship between derived temperature and pump energy at  $P = 0.5$  MPa,  
317  $\beta = 0.6$  and  $\phi = 1.0$ . As shown in Fig. 6, the derived temperature increases linearly with an increase in  
318 incident laser energy. By applying the linear extrapolation, the derived temperature with no

319 temperature rise can be determined as the intercept value of longitudinal axis. The *measured* resulting  
 320 temperature slope for the condition where  $P = 0.5$  MPa,  $\beta = 0.6$  and  $\phi = 1.0$  was 34.0 K/mJ. This large  
 321 sensitivity arises because of the highly focused beam used to enhance signal and improve spatial  
 322 resolution.

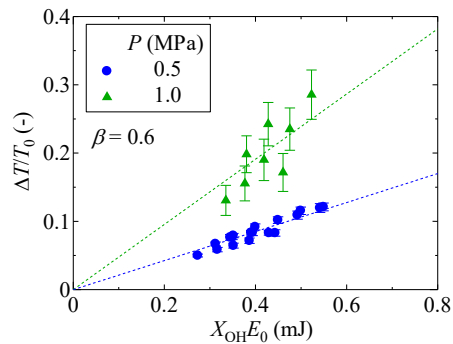
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325 *Figure 6. Relationship between apparent measured temperature,  $T$ , and pump laser energy,  $E_0$ .*

326



327

328 *Figure 7. Relationship between normalized temperature rise by incident laser absorption,*

329  *$\Delta T/T_0$ , and the product  $X_{OH}E_0$ .*

330

331 Although ideally this should be done for all cases, only a few conditions were evaluated over a  
 332 range of energies. In the case for  $P = 0.3$  MPa, the LITGS signal was relatively weak and it was not  
 333 possible to draw an appropriate curve. Figure 7 shows the general relationship between normalized  
 334 temperature rise,  $\Delta T/T_0$ , and the product of mole fraction of OH and incident laser energy,  $X_{OH}E_0$ , for  
 335 a range of energies and mole fractions. Here, all examined equivalence ratio cases that the incident

336 laser power varied (i.e.,  $\phi = 0.6, 0.8, 1.0, 1.2$  at  $P = 0.5$  MPa;  $\phi = 0.7$  and  $0.8$  at  $P = 1.0$  MPa) were  
337 plotted. The value of  $T_0$  was determined from the extrapolation of the relationship between  $T$  and  $E_0$ .  
338 The temperature rise is also influenced by the molecular composition at the measurement point [12]  
339 which in turn depends on the operating condition. However, as shown in Fig. 7, the variation of  $\Delta T/T_0$   
340 with  $E_0$  is linear in the range of energies considered, as expected from Eq. (5). The pressure effect is  
341 cancelled in Eq. (5) but the slope of  $\Delta T/T_0$  depends on pressure as shown in Fig. 7. Here we only  
342 discuss the influence on incident laser energy and mole fraction of the absorbing species, but the rate  
343 of thermalization via collisions can also affect the derived temperature [13].

344 In the case at  $P = 0.5$  MPa, the derived temperature agrees within 1.6% of the calculated adiabatic  
345 non-strained flame temperature (Fig. 5), with a calculated standard deviation of about 160 K at  $P =$   
346 0.5 MPa. We conclude that the temperature correction procedure must be systematically performed  
347 for accurate temperature evaluation. In the case at  $P = 1.0$  MPa, the corrected temperatures appear to  
348 be lower than the calculated adiabatic values but the derived temperature is close to the temperature at  
349 0.1 MPa obtained by OH-PLIF [21], suggesting that this may be the true value, and loss mechanisms  
350 may account for the discrepancy.

351

## 352 **5. Conclusions**

353 The study has demonstrated that LITGS temperatures can be successfully obtained using OH as  
354 the absorber in oxygen-enriched  $\text{CH}_4/\text{O}_2/\text{N}_2$  flames up to 1.0 MPa and  $\text{CH}_4/\text{O}_2$  flames at 0.3 MPa. The  
355 burned gas temperature can be derived from the LITGS signal, even for the mixtures where the flame  
356 temperature is as high as 3000 K. Therefore, the technique holds good potential for LITGS  
357 measurements under high pressure and high temperature flames, such as rocket combustion. Accurate  
358 measurements of temperature require calibration of the grating spacing, and correction for the  
359 temperature rise due to species absorption at high products of concentration and laser fluence. A  
360 procedure for temperature correction based on the linear extrapolation to zero absorption is  
361 recommended to minimize uncertainties.



362

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406

407 **Figure captions**

408 **Fig. 1** Experimental setup and the layout of the optical geometry through the high pressure  
409 chamber for high pressure OH-LITGS experiments. The symbols M, BS, BD, F and PMT  
410 represent mirror, beam splitter, beam dump, interference filter and photo multiplier tube,  
411 respectively.

412 **Fig. 2** Schematic figures of the modified burner nozzle for the calibration burner developed by  
413 Takeuchi et al. [21]: (a) Top-view; (b) Cross-sectional-view.

414 **Fig. 3** (a) Flame image of direct photo and (b) instantaneous OH-PLIF image at  $P = 1.0$  MPa for  $\phi$   
415  $= 1.0$  and  $\beta = 0.6$ .

416 **Fig. 4** Representative single shot LITGS signal for the equivalence ratio of  $\phi = 0.6$ : (a)  $P = 0.3$   
417 MPa,  $\beta = 0.6$ ; (b)  $P = 0.3$  MPa,  $\beta = 1.0$ ; (c)  $P = 0.5$  MPa,  $\beta = 0.6$ ; (d)  $P = 1.0$  MPa,  $\beta = 0.6$ .

418 **Fig. 5** Derived temperature from the LITGS signals with and without corrections (see text): (a)  $P =$   
419  $0.3$  MPa,  $\beta = 0.6$ ; (b)  $P = 0.3$  MPa,  $\beta = 1.0$ ; (c)  $P = 0.5$  MPa,  $\beta = 0.6$ ; (d)  $P = 1.0$  MPa,  $\beta =$   
420  $0.6$ . Solid lines are obtained from unstrained flame simulations.

421 **Fig. 6** Relationship between apparent measured temperature,  $T$ , and pump laser energy,  $E_0$ .

422 **Fig. 7** Relationship between normalized temperature rise by incident laser absorption,  $\Delta T/T_0$ , and  
423 the product  $X_{OH}E_0$ .