



Title	Turbulent burning velocity of ammonia/oxygen/nitrogen premixed flame in O ₂ -enriched air condition
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1 Turbulent burning velocity of ammonia/oxygen/nitrogen premixed flame in O₂-enriched air

2 condition

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12 Abstract

13 Ammonia is a promising hydrogen-energy carrier as well as a carbon-free fuel. However, turbulent
14 burning behavior of ammonia flame had yet to be sufficiently studied. In this work, laminar and
15 turbulent burning velocities of ammonia/oxygen/nitrogen flames were investigated under the
16 condition of oxygen enrichment. The turbulent burning velocity of ammonia/oxygen/nitrogen
17 mixtures was found to increase with increasing turbulence intensity. The ratio of the turbulent burning
18 velocity to stretched laminar burning velocity, U_{tr}/U_N , increased with the turbulence Karlovitz number.
19 However, because of the diffusional-thermal instability effect, given the same turbulent Karlovitz
20 numbers, U_{tr}/U_N in ammonia-lean cases is larger than in ammonia-rich cases. These findings indicate
21 that consideration of the effects of diffusional-thermal instability and of the turbulence is important
22 for the prediction of turbulent flame propagation velocity in ammonia combustion fields.

24 **Keywords:** Premixed ammonia/oxygen/nitrogen flame; Turbulent burning velocity; Laminar burning
25 velocity; O₂-enriched combustion

26

27

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29

30 **1. Introduction**

31 Given the increasing concerns about environmental impact from power generation by conventional
32 fuels, the search for alternative fuels has become increasingly important. Use of alternative fuels
33 produced from renewable sources can reduce greenhouse gases and pollutant emissions [1]. Ammonia
34 is considered to be one of the most-promising hydrogen-energy-carrier candidates and is an
35 independent alternative to hydrocarbon-based fuels [2]. The combustion of NH₃ does not release CO₂,
36 SO_x and soot [3–5]. Moreover, ammonia can be synthesized from renewable hydrogen and nitrogen
37 separated from air [6]. Since the thermal properties of ammonia are similar to those of propane in
38 terms of boiling temperature and liquefaction pressure, it is economical to be stored in large quantities
39 and transported in liquid-form [7].

40 However, despite the merits associated with ammonia as a fuel, there are many challenges in
41 ammonia combustion (such as high auto-ignition temperature (903 K), low flammability, high NO_x
42 emission, and low combustion intensity), which make it difficult to use in various energy devices
43 [8,9]. Furthermore, the low combustion intensity of the flame from an ammonia/air mixture makes
44 flame enhancement important for its application as a fuel. Various methods have been proposed to
45 enhance ammonia combustion. Hydrogen addition is a reasonable way of achieving environmentally
46 friendly flame enhancement. The relationship between laminar burning velocity and hydrogen
47 fraction in an ammonia/hydrogen binary fuel was revealed by Ichikawa et al. [10], Lee et al. [5], Li
48 et al. [11] and Kumar et al.[12]. In addition, blending hydrocarbon fuels with ammonia is also
49 important from the perspective of reducing greenhouse-gas emissions. Henshaw et al. [13] and Okafor
50 et al. [14] measured the CH₄/NH₃ mixture laminar burning velocity. Besides, ammonia combustion

51 under conditions of O₂ enrichment is also a promising way to enhance ammonia combustion due to
52 the thermal enhancement without changing the dominant chemistry. Such O₂-enriched combustion
53 enhances ammonia laminar burning velocity, a positive effect previously verified by Takeishi et
54 al.[15], Li et al. [2] and Mei et al.[16].

55 As reviewed above, laminar burning velocity is one of the most important parameters for
56 characterizing ammonia combustion behavior as it contains much physicochemical information. It
57 also is a reference parameter for many premixture flame phenomena, such as extinction, flashback,
58 blow off, and turbulent flame propagation [17]. On the other hand, turbulent flame is also important
59 for the practical use of fuels in combustor. Under turbulent vortices, the flame front is wrinkled, and
60 the flame surface area increases. Turbulence also induces strong, unsteady velocity gradients, which
61 modify the local transport of reactants and lead to local flame structures that depart significantly from
62 the generic one-dimensional flame structures [18]. Thus, a turbulent flame characteristically burns
63 faster than a laminar one. However, because of the weak intensity of premixed ammonia/air flames
64 and corresponding ease of being extinguished, which are due to their low laminar burning velocity
65 [19], most prior ammonia turbulent combustion research focused on the stabilization of ammonia
66 flames [20–23], while no research was conducted to reveal the fundamental turbulent burning
67 behavior of ammonia flame.

68 Therefore, using a constant-volume spherical chamber, we conducted experiments to study the
69 turbulent burning behavior of ammonia flame under the condition of O₂ enrichment and thus reveal
70 the fundamental turbulent combustion characteristics of the ammonia/oxygen/nitrogen premixture.

71 **2. Experimental setup and methods**

72 **2.1 Experimental apparatus**

73 The schematic of the experimental setup is outlined in Fig. 1. The ammonia combustion
74 experiments were carried out by using a constant-volume spherical chamber with a diameter of 200
75 mm and a height of 280 mm. The chamber's total volume was $6.19 \times 10^{-3} \text{ m}^3$.

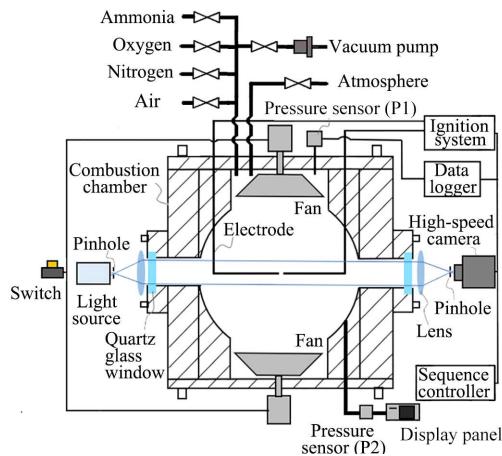


Fig. 1 Schematic of experimental apparatus.

76 A spark ignitor composed of two 2.0-mm-diam stainless-steel electrodes was employed in present
77 research. The gap between the two electrodes' sharp tips was set to 2 mm. A capacitor discharge
78 ignition (CDI) circuit system was connected to the spark electrode assembly to ignite the mixture. A
79 series of capacitors with a total capacitance of $50 \mu\text{F}$ was charged by 469 volts direct current in the
80 CDI circuit. The charged-voltage error was within $\pm 1.5\%$. The electrostatic energy, charged in the
81 capacitor circuit, was 5.5 J.

82 Turbulence was generated in the combustion chamber by two identical counter-rotation seven-
83 bladed fans mounted vertically and symmetrically inside the chamber. Each fan was rotated by
84 directly coupled electric motors (Maxon Motor, RE50) with separate speed controllers (Maxon Motor,

85 ESCON 50/5). The fans were precisely adjustable between 0 and 15,000 rpm. The fan rotation speed
86 error is within ± 1 rpm. Turbulence parameters were measured by particle image velocimetry (PIV)
87 measurement, as detailed in previous research [23]. In the center of the vessel that corresponds to the
88 region of optical access, the turbulence was found to be isotropic and homogeneous without regular
89 bulk motion. Turbulence intensity, u' , was proportional to the fan speed [24], found to be represented
90 by Eq. (1):

$$91 \quad u' = 0.00129 f_s \text{ m/s}, \quad (1)$$

92 where f_s is the fan speed in rpm. The longitudinal integral length scale, L_f , determined by two-
93 point correlation, was determined to be 20.9 mm regardless of the turbulence intensity [23].

94 Four optical-quartz glass observation windows were installed opposite each other in the chamber.
95 Flame propagation behavior was observed and recorded by schlieren photography, while the pressure
96 increase inside the chamber during flame propagation was measured with a pressure sensor (Valcom,
97 VPRTF-A4-(-0.1–2MPa) S-5, P1) and recorded by pressure transducer (GRAPHTEC, GL900) at 10
98 μs intervals. Schlieren photography was taken by a high-speed camera (Phantom, Miro C210) with
99 an LED light source (Hayashi-repic co., ltd., LA-HDF5010C). A pinhole was fixed in front of the
100 light source to form the spotlight effect. The pixel resolution of the camera was set to 576×576 with
101 a frame rate of 3000 fps.

102

103 **2.2 Experimental methods**

104 The experimental conditions are summarized in Table 1. In all the experiments, the initial pressure

105 and temperature were 0.1 MPa and 298 K. The temperature variations of the mixtures were within ± 3
 106 K. The turbulence intensity varied as 0–1.29 m/s. The composition of ammonia/oxygen/nitrogen
 107 mixture was defined by the equivalence ratio, ϕ , varied as 0.6–1.6, and was calculated on the basis
 108 of the ratio of ammonia to oxygen in the gas mixture [25] by Eq. (2):

$$109 \quad \phi = \frac{(O/F)_{stoic}}{O/F} = \frac{F/O}{(F/O)_{stoic}}, \quad (2)$$

110 where F represents mole of ammonia, and O represents mole of oxygen in the premixed gas mixture.

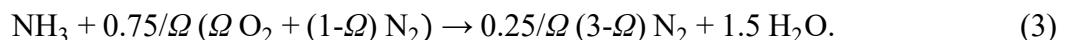
111

112 Table 1
 113 Experimental conditions of ammonia/oxygen/nitrogen premixture combustion

Experiment type	Ω	Equivalence ratio, ϕ	Turbulence intensity, u' (m/s)
Laminar	0.35	0.6, 0.8, 0.9, 1.0, 1.1, 1.2, 1.4	-
	0.4	0.6, 0.8, 0.9, 1.0, 1.1, 1.2, 1.4, 1.6	
Turbulent	0.4		0.32, 0.65, 0.97, 1.29

114

115 The chemical equation for ammonia/oxygen/nitrogen combustion under stoichiometric condition
 116 is given by Eq. (3):



118 Ω is expressed by Eq. (4):

$$119 \quad \Omega = \frac{X_{\text{O}_2}}{X_{\text{O}_2} + X_{\text{N}_2}}, \quad (4)$$

120 where X represents mole fraction. Taking consideration of the power density of ammonia flame and
 121 minimizing buoyancy effects [16,23,26], oxidizers with Ω of 0.35 and 0.4 were used for the research.

122 Laminar experiments under $\mathcal{Q} = 0.35$ were performed to verify the setup suitable for O₂-enriched
123 ammonia combustion experiments through comparing with the results of Mei et al. [16]. Experiments
124 on turbulent condition were carried out under $\mathcal{Q} = 0.4$. The premixed gases had composition ratios
125 based on their partial pressures, as measured with a pressure sensor (Valcom, VPRTF-A4-(-0.1–
126 1MPa)-5, P2). The maximum errors of NH₃ concentration, O₂ concentration, and pressure inside the
127 chamber were 1.5%, 1.5%, 5%, respectively. After filling the ammonia/oxygen/nitrogen mixtures in
128 the chamber, an isotropic mixture was formed by running the top and bottom fans. In laminar
129 experiments, the mixtures were ignited after 5 minutes of finishing stirring. For turbulence
130 experiments, two fans were kept running during experiments. At least six experiments were
131 performed for each condition to ensure repeatability of the results.

132 Figure 2 shows the pressure history inside the combustion chamber for $\phi = 0.6$ and $\phi = 1.0$ at
133 $u' = 1.23$ m/s. The observation time range differed depending on the equivalence ratio and turbulence
134 intensity. For $\phi = 0.6$, the pressure was constant at $\pm 5\%$ of ambient pressure during the observation
135 range (0–15 ms), whereas for $\phi = 1.0$, the observation range was lower because of the greater flame
136 propagation velocity. Overall, there was no pressure rise in the chamber during the observation range
137 for all conditions.

138

139

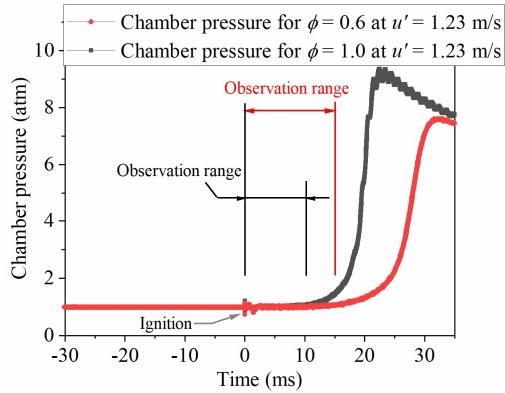


Fig. 2 Pressure history in chamber during combustion for $\phi = 0.6$ and 1.0 at $u' = 1.23$ m/s.

140 **3. Evaluation of burning velocity and ammonia/oxygen/nitrogen mixture properties**

141 **3.1 Evaluation of unstretched laminar burning velocity**

142 Unstretched laminar burning velocity was derived from measuring the spherical propagation flame
 143 images obtained by the high-speed camera. The flame propagation velocity, S_n , was calculated from
 144 the mean flame radius, r_{sch} , of a circle whose area was equal to area of the schlieren image of spherical
 145 flame against time by Eq. (5):

$$146 \quad S_n = \frac{dr_{sch}}{dt}. \quad (5)$$

147 During the laminar flame propagation process, the effects of the flame stretch must be considered,
 148 given that the spherically propagating flame had curvature. The flame stretch rate, ε , acting on the
 149 flame is defined [26–28] as Eq. (6):

$$150 \quad \varepsilon = \frac{1}{Area} \cdot \frac{d(Area)}{dt} = \frac{2}{r_{sch}} \cdot \frac{dr_{sch}}{dt}, \quad (6)$$

151 where $Area$ was the area of the spherical propagation flame front. Owing to the effect of diffusional–
 152 thermal instability, for non-unity Lewis number cases, the flame temperature and the laminar flame

153 propagation velocity were different from those in the unstretched case. The difference between
154 stretched laminar flame propagation velocity, S_n , and unstretched laminar flame propagation velocity,
155 S_S , can be expressed as a proportional relationship between burned gas Markstein length, L_b , and
156 stretched rate, ε , as expressed [29] by Eq. (7):

157
$$S_S - S_n = L_b \cdot \varepsilon. \quad (7)$$

158 By extrapolating the stretched laminar flame propagation velocity to an infinite flame radius, which
159 is equivalent to setting the flame stretched rate to 0, the unstretched laminar flame propagation
160 velocity can be obtained. The unstretched laminar burning velocity, U_L , was derived from Eq. (8):

161
$$U_L = \frac{\rho_b}{\rho_u} \cdot S_S. \quad (8)$$

162 The stretched laminar burning velocity, U_N , was calculated by Eq. (9):

163
$$U_N = \frac{\rho_b}{\rho_u} \cdot S_n, \quad (9)$$

164 where ρ_u and ρ_b are unburned mixture density and burned mixture density, respectively.

165 Kelly et al. [30] found that outwardly propagating spherical flames in a constant-volume chamber
166 may exhibit nonlinear relationship between the stretched flame propagation velocity, S_n , and stretched
167 rate, ε . To extrapolate the unstretched laminar propagation velocity and the burned gas Markstein
168 length, they proposed the nonlinear relationship expressed by Eq. (10):

169
$$\left(\frac{S_n}{S_S}\right)^2 \ln \left(\frac{S_n}{S_S}\right)^2 = -2 \frac{L_b \varepsilon}{S_S}. \quad (10)$$

170 The unstretched laminar flame propagation velocity, S_S , and the burned gas Markstein number, L_b ,

171 can be measured from linear extrapolation of $\ln(S_n)$ with ε/S_n^2 [14,31]. Okafor et al. [14] concluded
172 that the nonlinear equation was also appropriate for mixtures with Lewis numbers close to unity. In
173 our research, both linear and nonlinear equations were used to obtain the unstretched laminar burning
174 velocity, U_L , and the burned gas Markstein length, L_b .

175

176 **3.2 Evaluation of turbulent burning velocity**

177 The turbulent burning velocity is expressed by Eq. (11), the same as for the laminar burning
178 velocity [27,32,33]:

$$179 \quad U_{tr} = \frac{\rho_b}{\rho_u} \cdot \frac{dr_{sch}}{dt}. \quad (11)$$

180 In the case of turbulent flames, the flame is deformed by turbulent eddies. Because the schlieren
181 image is integrated along the light path, it is impossible to define the three-dimensionally wrinkled
182 flame front by using the schlieren photography. However, Bradly and Kitagawa et al. [32] revealed
183 that such burning velocity could be correlated to the turbulent burning velocity by planer Mie-
184 scattered imagery, and they also came to an important conclusion that the turbulent burning velocity
185 was associated with the entrainment and mass-consumption rate of the unburned mixture.

186

187 **3.3 Ammonia/oxygen/nitrogen mixture properties**

188 Among the properties of the ammonia/oxygen/nitrogen mixtures (Table 2), ρ_b and ρ_u were
189 burned and unburned gas mixture densities were obtained from the Chemical Equilibrium Application
190 (CEA) computer program of the U.S. National Aeronautics and Space Administration [34]. λ , c_p , α ,

191 ν and Le were thermal conductivity, specific heat at constant pressure, thermal diffusivity,
192 kinematic viscosity, and the Lewis number which were calculated using the website of the Dandy
193 research group at Colorado State University [35]. The Lewis number is defined [36] by Eq. (12):

194
$$Le_i = \frac{\alpha}{D_{i/N_2}}, \quad (12)$$

195 where D_{i/N_2} is the mass diffusivity of the deficient species “i”.

196 The effective Lewis number, Le_{eff} , of a single component fuel and an oxidizer can be expressed [37]
197 by Eq. (13):

198

199
$$Le_{eff} = I + \frac{(Le_E - I) + (Le_D - I)A}{I + A}. \quad (13)$$

200

201 In this equation, the Lewis number Le_E and Le_D are those defined based on the reactant that is
202 relatively in excess and deficient, respectively. A represent a measure of the mixture’s strength which
203 can be calculated by Eq. (14):

204
$$A = I + Ze(\Phi - I). \quad (14)$$

205 The constant Φ , which is always >1 , is defined as the mass ratio of excess-to-deficient reactants
206 in the mixture relative to their stoichiometric ratio [37,38]. Ze is Zel’dovich number which can be
207 expressed by Eq. (15):

208

209
$$Ze = E_a(T_{ad} - T_w)/(R^0 T_{ad}^2), \quad (15)$$

210

211 where R_0 is the universal gas constant, E_a is the activation energy which can be calculated [28,39] by

212 Eq. (16):

213

214

$$\frac{E_a}{R_0} = -2 \frac{\partial \ln(\rho_u U_L)}{\partial (1/T_{ad})}. \quad (16)$$

215

216 As shown in Table 2, although the equivalence ratio changes greatly from 0.6 to 1.6 under oxygen
217 content of 0.4, values of the effective Lewis number which change from 0.936 to 1.084, are very close
218 to unity. Thus, the differential diffusion effect (characterized by the effective Lewis number) is
219 comparably small.

220

221 Table 2

222 Properties of ammonia/oxygen/nitrogen mixtures [=, can't be defined]

Ω	ϕ	ρ_u [kg/m ³]	ρ_b [kg/m ³]	λ [10 ⁻³ W/m/K]	c_p [J/kg/K]	α [10 ⁻⁵ m ² /s]	v [10 ⁻⁵ m ² /s]	Le	T _{ad} [K]	Le_{eff}
0.35	0.6	1.0908	0.14938	27.559	1144.6	2.2074	1.560	0.896	2062.14	0.942
	0.8	1.0650	0.12707	27.804	1204.4	2.2034	1.556	0.894	2324.47	0.948
	0.9	1.0527	0.11985	27.911	1204.4	2.2012	1.554	0.883	2412.55	0.965
	1.0	1.0410	0.11483	28.015	1224	2.1987	1.552	=	2460.04	1.000
	1.1	1.0304	0.11249	28.099	1241.5	2.1966	1.550	1.125	2444.05	1.034
	1.2	1.0198	0.11227	28.181	1259.4	2.1943	1.549	1.122	2377.48	1.052
	1.4	1.0002	0.11419	28.337	1294.3	2.1889	1.545	1.119	2293.83	1.070
	0.6	1.0861	0.13940	27.666	1155.2	2.2051	1.559	0.906	2182.39	0.936
0.4	0.8	1.0570	0.11972	27.922	1200.7	2.2002	1.554	0.88	2423.05	0.945
	0.9	1.0440	0.11340	28.030	1221.8	2.1975	1.552	0.868	2497.35	0.963
	1.0	1.0319	0.10893	28.128	1241.9	2.1948	1.550	=	2534.38	0.999
	1.1	1.0206	0.10646	28.216	1261.1	2.1921	1.548	1.138	2521.72	1.033
	1.2	1.0101	0.10575	28.296	1279.5	2.1895	1.546	1.135	2463.98	1.052
	1.4	0.9909	0.107	28.435	1313.8	2.1842	1.543	1.13	2296.3	1.072
	1.6	0.97397	0.10978	28.552	1345.3	2.1791	1.5396	1.125	2122.21	1.084

223

224 **4. Results and discussion**

225 **4.1 Flame observation**

226 Figure 3 shows the sequence of schlieren images of ammonia/oxygen/nitrogen flames from 1 to 6

227 ms for $\phi = 0.6$ under laminar and four different turbulence intensity cases. It is apparent that as the
 228 turbulent flame expands outwardly, the flame becomes more wrinkled and accelerated. In the initial
 229 period of turbulent flame propagation, only eddies of a size smaller than the flame size could wrinkle
 230 the flame front, but with the development of the flame radius, the flame can be affected by wider
 231 range of turbulence wavelength. Also, Kolmogorov and Taylor's length scales become smaller as the
 232 turbulence intensity increases. Therefore, as the turbulence intensity increases, more smaller patterns
 233 of flame front deformations can be seen in Fig. 3. Also, the nonuniformity of the wrinkling effects of
 234 eddies increases with increasing turbulence intensity [40,41]. Accordingly, the flame propagates
 235 faster with increasing turbulence intensity.

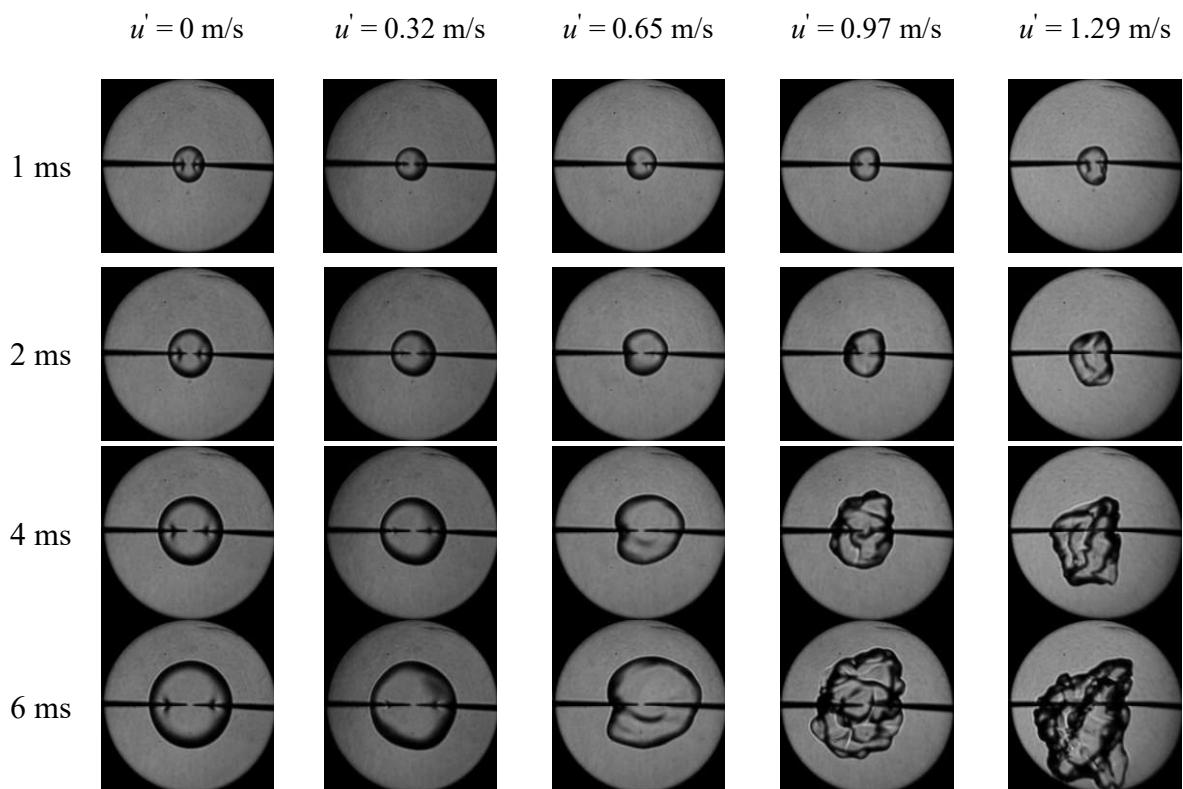


Fig. 3 Schlieren images of ammonia/oxygen/nitrogen mixtures under $\phi = 0.6$ at various turbulence intensities.

236 The 16-bit images were processed by gray levels from 0 = black (flame) to 255 = white

237 (background). The threshold value was selected based on visual inspection. The range of the threshold
238 value used for processing the images was from 50 to 100 depending on the obtained images. The
239 threshold-value effect on the radius and velocity deviations was within $\pm 2\%$ (see Appendix). Then
240 the processed image was transferred to the binary image in which the flame was black and background
241 was white. Subsequently, the flame area was calculated by the number of included pixels of the flame
242 area and the mean flame radius, r_{sch} , was determined by the radius of the equivalent circle area under
243 both laminar and turbulent conditions.

244 Figure 4 illustrates the flame radius as a function of elapsed time under $\phi = 0.6$ for various
245 turbulence intensities. It is apparent that at the beginning, the flame propagation was affected by the
246 ignition energy, but after the flame developed beyond the ignition affected period, under the same
247 elapsed time, the flame radius increased with the increase of turbulence intensity.

248

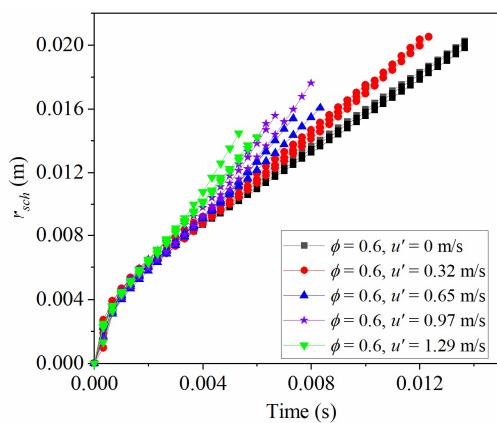


Fig. 4 Flame radius as function of time
under $\phi = 0.6$ at various turbulence
intensities.

249

250 4.2 Unstretched laminar burning velocity of the ammonia/oxygen/nitrogen flame

251 The ignition energy effect is checked by changing ignition energy from 0.3 to 5.5 J, as shown in

252 Fig. 5. The critical flame radius, which was used to eliminate the ignition energy affected period,
 253 observed by ammonia/oxygen/nitrogen combustion for $\phi = 1.0$ at $\Omega = 0.4$, was ~ 0.008 m. (This
 254 method was used by Chen et al.[42] and Huang et al. [43].) So, the radius that was < 0.008 m was not
 255 counted in laminar and turbulent studies.

256

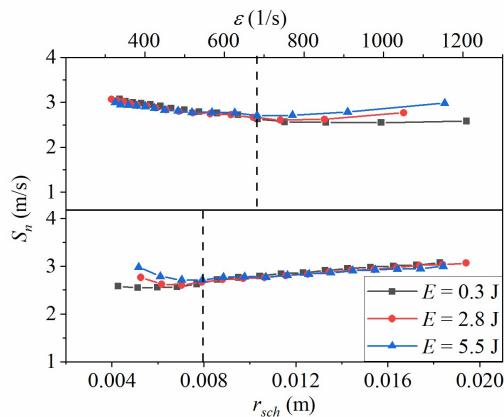


Fig. 5 Stretched laminar flame propagation velocity as functions of flame radius and stretch rate for ammonia/oxygen/nitrogen mixture at $\phi = 1.0$ on $\Omega = 0.4$.

257 In our research, the outwardly propagating spherical flame (OPF) in the constant-pressure method
 258 was used to obtain the unstretched laminar burning velocity. Chen et al. [44] had noted that the
 259 ignition energy, confinement, buoyancy, flame instability, and nonlinearity of the flame speed-stretch
 260 rate relationship can cause inaccuracy of unstretched laminar flame speed measurement. The relation
 261 between the stretched laminar flame propagation velocity, S_n , and the flame stretch rate, ϵ , under
 262 conditions of $\phi = 0.8$ and $\phi = 1.2$ are shown in Fig. 6. According to Kelley [30], flame propagation
 263 in a constant-pressure environment can be divided into three distinctive periods: an ignition energy
 264 affected period, a quasi-steady period, and a chamber confinement affected period. The ignition
 265 energy effected period was determined by the critical radius (Fig. 5). The confinement effect was
 266 neglected because the observation period was relatively small compared with chamber volume. Mei

267 et al. [16] pointed out that the buoyancy effect on the ammonia/oxygen/nitrogen flame was almost
268 eliminated under oxygen contents of 35% and 45% because flames for both were nearly spherical.
269 The same phenomenon was found in present research. Also, the buoyancy effect was ignored because
270 there was no noticeable change in the flame symmetry under any tested conditions. Too, because the
271 flames within the observation range in the present study were stable, there was no uncertainty caused
272 by instability. Additionally, linear and nonlinear methods were carefully applied to avoid the
273 uncertainties induced by the nonlinearity of the flame speed-stretch rate relationship.

274

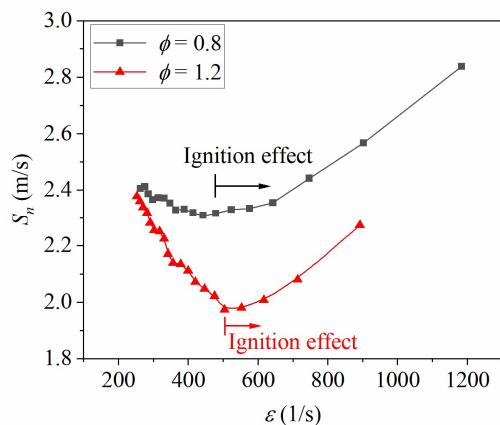


Fig. 6 The unstretched laminar propagation velocity as a function of stretch rate

275

276 Figure 7 shows the unstretched laminar burning velocities of the ammonia/oxygen/nitrogen
277 premixture flame, which were obtained by using Eqs. (7) and (10). The results obtained by Mei et al.
278 [16] by the same experimental method for $\Omega = 0.35$ were similar to those from our current research.
279 Accordingly, the reproducibility of the flame propagation experiments of ammonia O₂-enriched
280 combustion was confirmed.

281

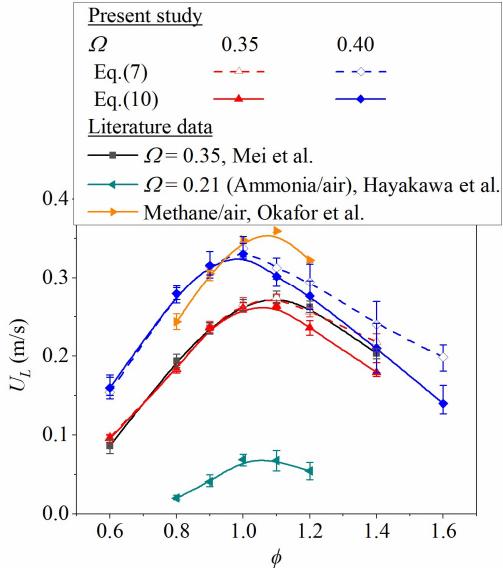


Fig. 7 Experimental results of correlation of unstretched laminar burning velocity with equivalence ratio under various oxygen contents in oxidizer

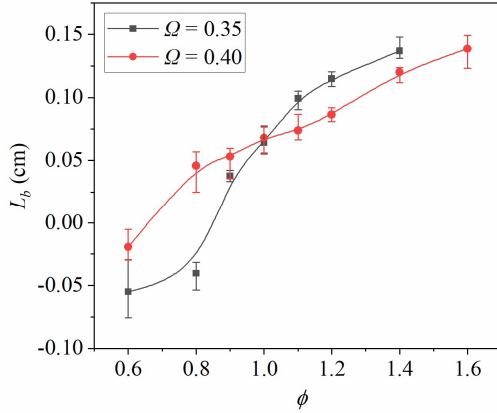
282 As indicated in Fig. 7, under the same ammonia/oxygen/nitrogen equivalence ratio, the unstretched
 283 laminar burning velocity of the ammonia/oxygen/nitrogen mixture increases with increasing Ω from
 284 0.21 to 0.4. The maximum laminar burning velocity was 0.337 m/s at $\phi = 1.0$ under $\Omega = 0.4$ which
 285 was approximately 4.9 times the value under the condition $\Omega = 0.21$. From comparing with laminar
 286 burning velocity of CH₄/air [2], 40% oxygen content was found to enhance the ammonia laminar
 287 burning velocity to the level of methane/air laminar burning velocity.

288 On the other hand, the unstretched laminar burning velocities obtained from linear and nonlinear
 289 equations are approximately same on ammonia-lean and under stoichiometric conditions. However,
 290 the disparity increased with the rise of the equivalence ratio in ammonia-rich cases. The disparity
 291 between the two methods was considered by Okafor et al. [14] to have been caused by the increasing
 292 sensitivity of the flame to stretch.

293 The sensitivity of the laminar burning velocity to the flame stretch rate can be evaluated by the

294 burned gas Markstein length, L_b . Fig. 8 shows the burned gas Markstein length which was obtained
 295 by the non-linear equation. Additionally, L_b increases with increase of ammonia equivalence ratio for
 296 both $\Omega = 0.35$ and 0.4 cases.

297



298 Fig. 8 Variation of the burned gas Markstein length with the equivalence ratio and oxygen content

299 From the asymptotic analysis of the premixed flame structure, L_b can be expressed [14,29] by Eq.
 300 (17):

301

302
$$L_b = \delta_F [f_1(\sigma) + Ze(Le-1)f_2(\sigma)], \quad (17)$$

303

304 where the $f_1(\sigma)$ and $f_2(\sigma)$ are functions of the thermal-expansion coefficient and are always
 305 positive for realistic coefficient values of σ [14]. In present research, the burned gas Markstein
 306 length was dominated by the Zel'dovich number, flame thickness, and Lewis number. Flame
 307 thickness, δ_F , is calculate [45] by Eq. (18):

308

$$\delta_F = \frac{\lambda}{c_p \rho_u U_L}. \quad (18)$$

309 Figure 9 shows the Ze and δ_F under different cases. Because Ze promotes the effect of a non-unity
 310 Lewis number, the effect of an increase in Ze on the sensitivity of the flames to stretch depends on

311 the value of the Lewis number [14]. In ammonia-lean cases in which $\text{Le} < 1$, Ze decreases with
 312 increasing L_b , while Ze increases with increasing L_b in ammonia-rich cases.

313

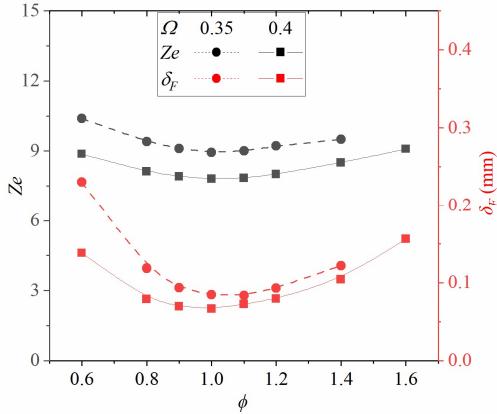


Fig. 9 Flame thickness and Zel'dovich number under different equivalence ratios and oxygen contents.

314 **4.3 Turbulent flame structure of ammonia/oxygen/nitrogen mixture under conditions of oxygen
 315 enrichment**

316 To reveal the turbulence longitudinal integral length scale effect on the flame front, Borghi and
 317 Peters proposed four kinds of turbulent flame regimes [19]. As shown in Fig. 10, the Peters regime
 318 diagram indicates four flame regimes determined by the ratio of longitudinal integral length scale (L_f)
 319 to laminar flame thickness (δ_F), with the ratio of turbulence intensity (u') to unstretched laminar
 320 burning velocity (U_L), including the wrinkled flamelets regime, the corrugated flamelets regime, the
 321 thin reaction zone, and the broken reaction zone.

322

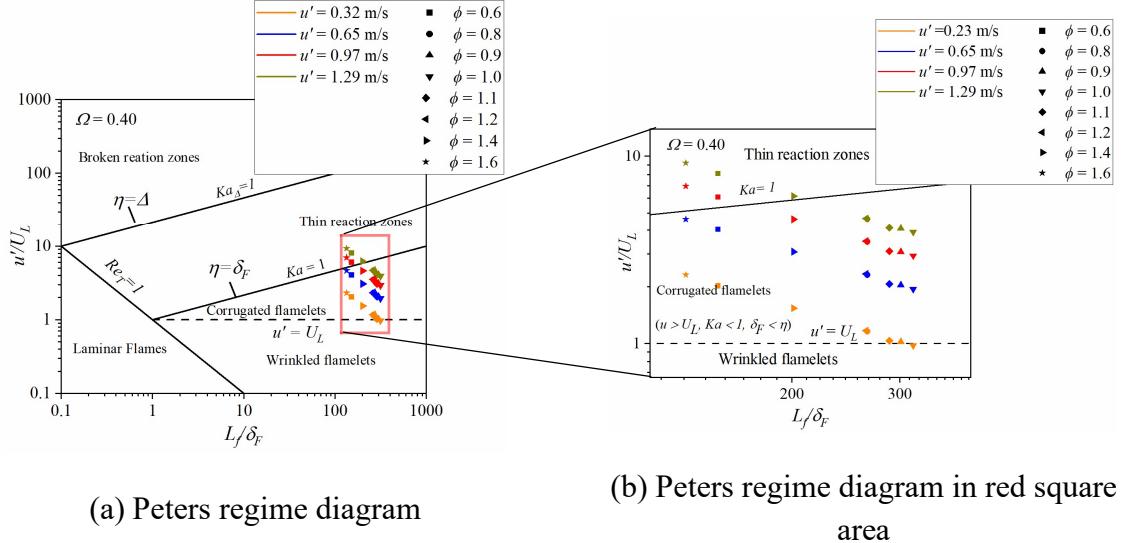


Fig. 10 Regimes of present experimental conditions on turbulent combustion diagram.

323

324 Most of the ammonia/oxygen/nitrogen turbulent flames were in the corrugated flamelets regime.
 325 In this regime, turbulent eddies push the flame front and undergo convolution, which increases the
 326 flame surface area (Fig.3) and the chemical reaction time is faster than the turbulent mixing. This
 327 means that the turbulent eddies can only wrinkle the laminar flamelets and cannot influence the
 328 chemical reaction within the flame front.

329 The turbulence Karlovitz number, which is the ratio of the chemical time scale to the time scale of
 330 turbulent eddies, can be calculated by Eq. (19) as a specific value of flame stretch caused by turbulent
 331 flow to turbulent flame [46]:

$$332 \quad Ka = \frac{\delta_F}{U_L} \sqrt{\frac{\lambda_f}{u'}}, \quad (19)$$

333 where U_L is the unstretched laminar burning velocity, λ_f is the longitudinal Taylor microscale of
 334 turbulent field, and u' is turbulence intensity, δ_F is the thickness of preheat zone.

335 As shown in Fig. 11, the turbulent Karlovitz number (Ka) increases with increasing turbulence

336 intensity irrespective of the equivalence ratio. Moreover, it decreases with increasing equivalence
 337 ratio in ammonia-lean cases but it increases with increasing equivalence ratio in ammonia-rich cases.
 338 However, all $Ka < 1$. Thus, compared to the time scale of small eddies, chemical reaction rates were
 339 faster enough that turbulence eddies just wrinkled the flamelets and did not affect the chemical
 340 reaction within the flame front.

341

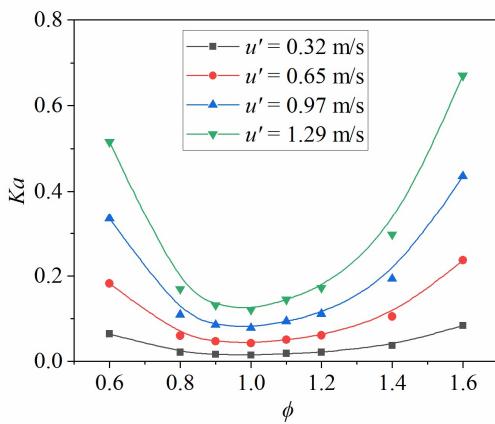


Fig. 11 Turbulence Karlovitz number (Ka) under different equivalence ratios and turbulence intensities.

342 **4.4 Turbulence burning velocity of ammonia/oxygen/nitrogen mixture under condition of**
 343 **oxygen enrichment**

344 Figure 12 shows the turbulent burning velocity U_{tr} and stretched laminar burning velocity U_N as
 345 functions of flame radius, r_{sch} , under various turbulence intensities at $\phi = 0.6$. As the flame radius
 346 increased from 0.004 m to 0.008 m, the turbulent burning velocity decreased because of the ignition
 347 energy effect. The ignition energy affected period, in which $r_{sch} < 0.008$ m, was ignored, but after the
 348 ignition energy affected period, the turbulent burning velocity increased with increasing flame radius
 349 for all turbulence intensity cases. For the same flame radius, the turbulent burning velocity increases
 350 with increasing turbulence intensity. The turbulent trends are the same for the other equivalence ratio
 351 cases.

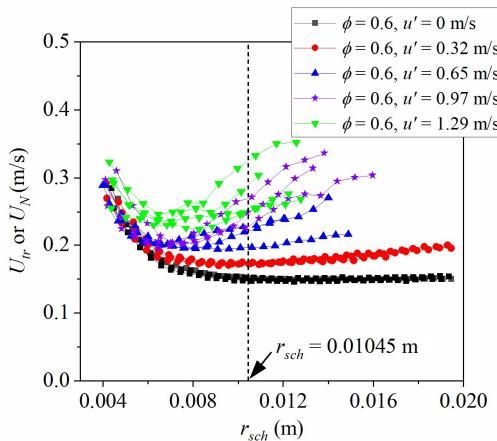


Fig. 12 Turbulent burning velocity developing as a function of flame radius for $\phi = 0.6$.

354 To examine the turbulence intensity effect on burning velocity, the burning velocity at flame
 355 diameter of the turbulence longitudinal integral length scale was used for comparison. The method
 356 has been used for studying premixed turbulent combustion of methane/air, hydrogen/methane/air,
 357 hydrogen/octane/air, hydrogen/air, iso-octane/air, methanol/air [24,47–52]. Moreover, for the laminar
 358 case, the stretched laminar burning velocity at flame diameter of the longitudinal integral length scale
 359 was used for comparison.

360 Figure 13 shows the turbulent burning velocity and stretched laminar burning velocity at flame
 361 diameter of L_f under various equivalence ratio, ϕ . It was found that turbulent burning velocity
 362 increases with increasing turbulence intensity. The increase of turbulent burning velocity of the
 363 ammonia/oxygen/nitrogen mixture is caused by the increase of the turbulent flame surface area
 364 deformed by turbulent eddies. Maximum turbulent burning velocity is at $\phi = 1.0$, the same as for
 365 unstretched laminar burning velocity. However, as shown in Fig. 13, somewhat large scatter is
 366 observed under conditions of high turbulence intensity. One reason for the large scatter is that the

367 flame is highly deformed at relative high turbulence intensities. This phenomenon has also been
 368 observed in other instances of gaseous fuel turbulent flame propagation [53,54]. A second reason for
 369 the scatter is that the effects of eddies on flame propagation increase with increasing turbulence
 370 intensity. There are turbulent eddies of various sizes in the turbulent flow field, and the wrinkling
 371 effects differ according to each eddy size [24]. Accordingly, the nonuniformity of wrinkling effects
 372 of eddies increases with increasing turbulence intensity.
 373

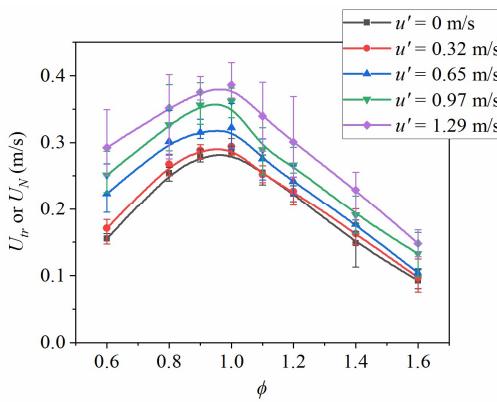


Fig. 13 Turbulent burning velocity and
 stretched laminar burning velocity at
 flame diameter of longitudinal integral
 length scale as a function of equivalence
 ratio.

374 Figure 14 shows the ratio of turbulent burning velocity to the stretched laminar burning velocity,
 375 U_{tr}/U_N , as a function of ϕ . Through using the ratio U_{tr}/U_N , the effect of the global stretch that appears
 376 in both the laminar and turbulent cases can be eliminated. Globally, the ratio U_{tr}/U_N decreases with
 377 increasing equivalence ratio for ammonia-lean cases but increases with increasing of equivalence
 378 ratio for ammonia-rich cases. Also, with increasing turbulence intensity, the ratio U_{tr}/U_N increases.
 379 To examine these tendencies, the turbulence Karlovitz number, Ka , was introduced and the
 380 relationship between turbulence Karlovitz number and the ratio of turbulent burning velocity to

381 stretched laminar burning velocity was studied.

382

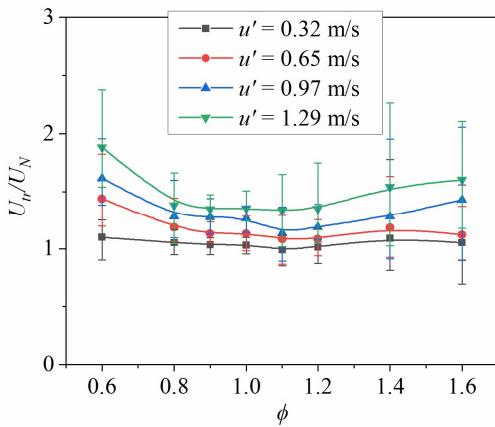


Fig. 14 The relation of the ratio of U_{tr}/U_N with ammonia equivalence ratio.

383 Figure 15(a) shows the relationship between turbulence Ka and U_{tr}/U_N . The ratio, U_{tr}/U_N , increases
384 with increasing turbulence Ka . As shown in Fig. 15(b), the ratio U_{tr}/U_N for ammonia-lean cases, is
385 higher than for ammonia-rich cases. This is most likely due to the effect of diffusional–thermal
386 instability. The local burning velocity in the convex structure increases when $Le < 1$, i.e., the mass
387 diffusion of reactants from the unburned area to the burned area excels the thermal diffusion from the
388 burned area to the unburned area, and a convex structure develops. On the other hand, the local
389 temperature and burning velocity in concave areas decrease, and a concave shape develops also.
390 Eventually, overall burning velocity increases due to the increase in flame surface area because of the
391 diffusional–thermal instability effect caused when $Le < 1$. Hence, turbulence eddies and the
392 diffusional–thermal instability effect induced higher turbulent burning velocity in ammonia-lean
393 cases under the same turbulence Ka . These findings indicate that not only the consideration of the
394 turbulence effect but also the consideration of the diffusional–thermal instability effect is important

395 to predict the turbulent burning velocity of ammonia flame. Consideration of the effect of the
 396 diffusional-thermal instability when simulating flame propagation velocity can contribute greatly to
 397 the future design of combustor using ammonia as a fuel.

398

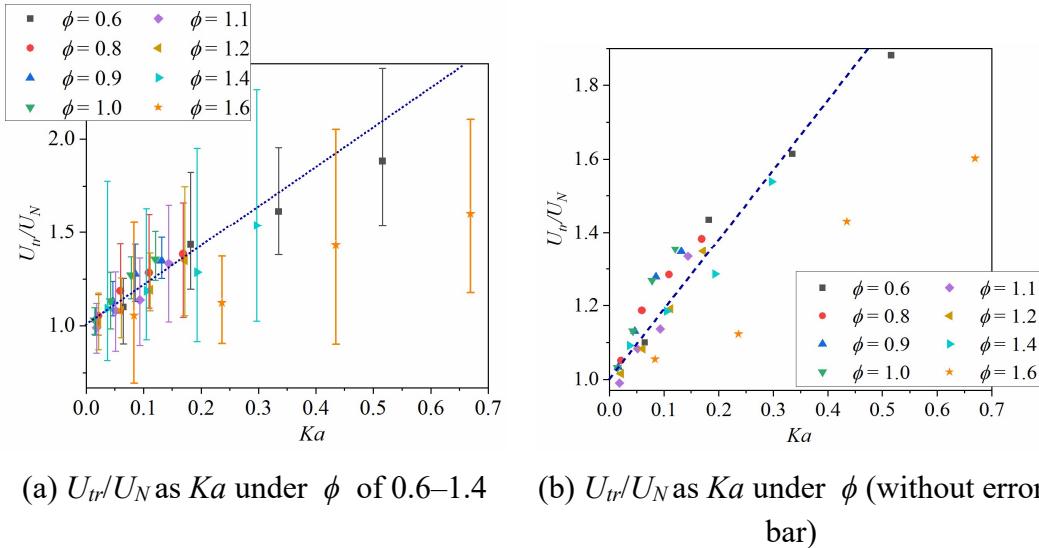


Fig. 15 Ratio of turbulent burning velocity to stretched laminar burning velocity (U_{tr}/U_N) in terms of turbulence Karlovitz number (Ka).

399

400 However, for the case of $\phi = 0.6$, the U_{tr}/U_N as the turbulence Ka is slightly lower than the average
 401 line. This is considered to be caused by the lower adiabatic flame temperature, T_{ad} , which is shown
 402 in Table 2. For the case when $\phi = 1.6$, the Lewis number is almost the same as when $\phi = 1.4$.
 403 However, because of lower T_{ad} , the ratio of U_{tr}/U_N under the condition of $\phi = 1.6$ is much smaller
 404 than that in the case when $\phi = 1.4$. Hence, under the condition of $\phi = 0.6$, the lower adiabatic flame
 405 temperature limits the effect of diffusional-thermal instability. There is a possibility that the local
 406 quenching happens owing to the lower adiabatic flame temperature even though the basic diffusional-
 407 thermal instability effect is strong for ammonia-lean flame. That is, the contribution of the effect of
 408 diffusional-thermal instability to increase flame surface area is limited by local quenching due to the

409 lower adiabatic flame temperature for the case when $\phi = 0.6$. Although the diffusional-thermal
410 instability effect is limited for the $\phi = 0.6$ case, it is still important because the ratio of U_{tr}/U_N under
411 $\phi = 0.6$ is much larger than that under $\phi = 1.6$ even though T_{ad} is almost the same.

412 **5. Conclusion**

413 The effect of turbulence intensity on the burning characteristics of an ammonia/oxygen/nitrogen
414 mixture was investigated under conditions of oxygen enrichment. The principal findings are as
415 follows:

416 1. The turbulent burning velocity of the ammonia/oxygen/nitrogen flame increased with the
417 increasing turbulence intensity owing to the increase of flame surface area (due to the increase
418 of turbulence intensity).

419 2. Under the same turbulence Karlovitz number, the ratio of turbulent burning velocity to
420 the stretched laminar burning velocity in ammonia-lean cases was larger than that in ammonia-
421 rich cases. This is caused by the effect of diffusional-thermal instability. Consideration of both
422 effects of the diffusional-thermal instability and the turbulence is important for the prediction
423 of turbulent flame propagation velocity in ammonia combustion fields.

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