Title	A Novel Characte Supercritical Diff	eristic Length of Detonation Relevant to Traction
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2 Abstract For stoichiometric  $C_2H_4$ – $O_2$  and  $C_2H_2$ – $O_2$  mixtures with or without argon dilution, the processes of 3 detonation diffraction have been investigated in a two-dimensional setup through high-speed schlieren imaging, 4 with the characteristic length and the stability of detonation varied by regulating the initial pressure and argon mole 5 fraction of the mixture. In particular, a length relevant to the process of supercritical diffraction (i.e., distance from 6 the channel end corner to reflection point of the transverse detonation on the channel end face, reflection point 7 distance in short) was deduced from obtained sequential schlieren images and analyzed. The reflection point 8 distance can be idealized for the infinitely wide donor channel, and thus it can be an parameter in which properties 9 intrinsic to each detonable mixture are manifested. Experimental results showed that the reflection point distance 10 was roughly inversely proportional to the initial pressure for identical mixtures and independent of the width of the 11 donor channel at high initial pressures. For a certain combination of the fuel and oxidizer, correlations between the 12 reflection point distance and the initial partial pressure of fuel were very similar regardless of the argon mole 13 fraction. Critical conditions of the diffraction problem could be given for the ratio of the reflection point distance to 14 the channel width, and it was suggested that the critical value lies in a range of 3–5 and does not significantly 15 depend on the stability of the mixture.

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Keywords Reflection point distance, Detonation dynamic parameters, Detonation diffraction, Critical tube
 diameter, Detonation stability, Schlieren imaging

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24		List of symbols
25	$D_{ m CJ}$	Chapman–Jouguet (CJ) velocity
26	$d_{\rm dt}$	Diameter of detonation tube
27	Ea	Activation energy
28	$p_0$	Initial pressure of mixture
29	$p_{0,\mathrm{f}}$	Initial partial pressure of fuel
30	lc	Channel width
31	$l_{ m r}$	Reflection point distance
32	R	Universal gas constant
33	$T_0$	Initial temperature
34	$T_{\rm vN}$	von Neumann temperature
35	t <sub>oc</sub>	Depth of observation chamber
36	УАг	Argon mole fraction
37	$\Delta_{\mathrm{i}}$	Induction length
38	λ	Detonation cell width
39	$\tau_{i}$	Induction time
40	$\tau_{\rm r}$	Reaction time

 $\chi$  Detonation stability parameter

# 1. Introduction

44	When a planar detonation wave propagating in a channel emerges into a wider space, the wave front
45	undergoes deformation and diffraction due to expansion of the flow behind the leading shock wave. The
46	transmission of the detonation from the channel into the wider space can fail under certain conditions. Based on
47	success or failure in the transmission, the diffraction process is often classified as supercritical or subcritical,
48	respectively [1], and there exists a critical condition for the size of the channel, depending on the configuration of
49	the channel and the composition and thermodynamic states of the detonable gas mixture [1, 2]. For instance,
50	Zel'dovich et al. [3] pointed out that a detonation wave in a cylindrical tube will fail in transmission upon
51	diffraction when the diameter of the tube is smaller than a certain value, which is called the critical tube diameter.
52	The critical tube diameter is a measure of the detonability of combustible mixtures [4, 5] and known as a
53	typical dynamic parameter of detonation phenomena, although quantitatively predictive theory for the critical tube
54	diameter is still under development [4]. The critical tube diameter has often been discussed in relation to the
55	detonation cell size in order to predict its value. However, the relationship between the critical tube diameter and
56	detonation cell width is not completely universal. The relationship depends on the geometry of the flow field and
57	the stability of the mixture. Mitrofanov and Soloukhin [6] discovered that, for the stoichiometric acetylene-oxygen

58	mixture at low pressures, the critical diameter is 13 or 10 times the cell width for a cylindrical tube where the flow
59	field is three-dimensional and a thin rectangular channel where the flow field is two-dimensional, respectively.
60	Later, the correlation between the critical tube diameter and cell width was confirmed for wider ranges of the initial
61	pressure and wider variety of fuel-oxidizer mixtures and channel geometry in other works [7-13]. This correlation,
62	however, was found not to hold for more stable mixtures with regular cellular patterns, including mixtures with
63	high argon dilution; instead, larger numbers of cells are required for a planar detonation wave to successfully
64	transmit from a channel into unconfined space [14-17]. Although the theoretical aspect of the correlation between
65	the critical channel size and the cell width remains an open problem, differences in the manner of the correlation
66	among mixtures with different stabilities are associated with the differences in the failure mechanisms of detonation
67	transmission [2, 4, 18–20].
68	Hence, many experiments and numerical simulations have been done to investigate the process of detonation
69	diffraction in detail. In an experiment by Bartlmä and Schröder [21], high-time-resolution sequential schlieren
70	photographs were obtained for processes of supercritical diffraction (success in transmission) and subcritical
71	diffraction (failure in transmission) using a multi-spark optical system. Based on the observations, they concluded
72	that re-initiation and formation of a transverse detonation are necessary for successful detonation transmission.
73	Jones et al. [22] performed two-dimensional numerical simulations to investigate the influences of the cellular
74	structure on the diffraction process. They concluded that transverse waves are necessary for re-initiation of the

75	detonation, and the number of the transverse waves are also important because successive propagation of the
76	transverse waves leads to a local explosion and subsequent transverse detonation. Pintgen and Shepherd [23]
77	examined the influence of the detonation stability on the features of diffraction through simultaneous schlieren
78	imaging, chemiluminescence imaging, and laser-induced fluorescence (LIF) imaging of OH radicals. In that study,
79	they found that, in subcritical diffraction processes, the detonation velocity on the symmetrical axis for a more
80	stable mixture (hydrogen-oxygen-argon mixture) decays significantly slower than for a more unstable mixture
81	(hydrogen-nitrous oxide mixture), whereas, in supercritical diffraction processes, the re-initiation events appeared
82	similar for the two mixtures. In recent years, Deiterding [24] and Li et al. [25] performed high-resolution numerical
83	simulations based on the inviscid, reactive Euler equations. In the simulations, cell-scale physics were resolved by
84	means of adaptive mesh refinement (AMR) and parallel computing, and the reproduced critical conditions between
85	channel width and cell size were quantitatively consistent with experiments. Mehrjoo et al. [26, 27] examined the
86	influence of cellular instability on detonation diffraction by perturbing the flow behind the leading shock with
87	obstacles or porous walled tubes. In the experiments, for more stable mixtures with high argon dilution, the critical
88	condition between the tube and cell sizes was not significantly affected, whereas for unstable mixtures, the critical
89	condition was significantly changed.
90	On the other hand, another correlation regarding the detonation diffraction problem is also becoming evident

91 for the reflection point distance of transverse detonation, which is the distance on a tube end face between the tube

92	end corner and a point where a detonation wave reattaches by re-initiation. Murray and Lee [28] discovered a
93	circular pattern on a soot foil on the tube end wall for equimolar acetylene-oxygen mixtures, and they found that
94	the distance between the circular pattern and the tube inner wall, which is equivalent to the reflection point distance,
95	was well correlated with detonation cell width. Nagura et al. [29, 30] also concluded that the reflection point
96	distance is roughly proportional to the detonation cell width for stoichiometric ethylene-oxygen and
97	acetylene-oxygen mixtures. These facts are exceptionally important, because if there exists some correlation
98	between reflection point distance and cell width, it would mean that reflection point distance, which can be easier
99	to measure, is equivalent to the cell width.
100	In the present study, for stoichiometric ethylene-oxygen and acetylene-oxygen gas mixtures, we investigated
101	processes of detonation diffraction through high-speed schlieren imaging in a two-dimensional setup with the initial
102	pressure and argon dilution ratio of the mixture varied. Based on the obtained sequential schlieren images, the
103	reflection point distance relevant to the supercritical diffraction process was deduced and examined as a
104	characteristic length of detonation.
105	
106	2. Experimental Setup
107	2.1. Experimental apparatus
108	The experimental apparatus consisted mainly of an observation chamber, detonation tube, dump tank, and

109 schlieren imaging system. A schematic of the observation chamber is shown in Fig. 1. The observation chamber

110	was connected to the detonation tube on the lower side, and with the dump tank in the downstream direction. In an
111	operation, a planar detonation wave generated in the detonation tube was introduced into the observation chamber
112	through an inlet channel from the lower side. The planar detonation wave then experienced an abrupt expansion at
113	the end of the inlet channel and was diffracted toward the upper right within the rectangular diffraction region. Note
114	that the flow-fields obtained in the present setup can be regarded as right halves of symmetric flow-fields in
115	cylindrical diffraction; the vertical wall of the observation chamber corresponds to the line of symmetry. Once the
116	detonation wave reached a diaphragm inserted at the joint between the observation chamber and the dump tank, the
117	diaphragm was ruptured, and the high temperature and pressure burnt gas was safely released into the dump tank.
118	During this sequence of events, the process of detonation diffraction occurring within the observation chamber was
119	visualized by the schlieren method and recorded with a high-speed video camera.
120	The observation chamber, consisting of inlet and outlet channels and a diffraction region, had a structure
121	uniform in depth between two parallel optical windows. The gap between the optical windows was $t_{oc} = 16$ mm,
122	which was significantly greater than the detonation cell widths at the conditions of interest, thus transverse waves
123	in depth were not suppressed. The inlet channel was 90-mm long and had a rectangular cross-section with a width
124	of $l_c = 10$ or 20 mm (variable) and a depth of $t_{oc}$ ; the aspect ratio of the cross-section was thus $t_{oc}/l_c = 1.6$ or 0.8,
125	respectively. However, regardless of the aspect ratio, the diffraction taking place in the present setup was
126	cylindrical (two-dimensional) but not spherical (three-dimensional) because the flow was bounded by the parallel

127	windows. Note that this situation is equivalent to the case of the infinite aspect ratio in unbounded, fully open space
128	[11]. The dimensions of the diffraction region were 60 mm in length, 80 mm in width, and $t_{oc}$ in depth, and the
129	depth is identical with the inlet channel. The detonation tube had a length of 560 mm and a circular cross-section
130	with an inner diameter of $d_{dt} = 25.8$ mm, and also was equipped with a Shchelkin spiral at the bottom. The
131	diaphragm separating the observation chamber from the dump tank was made of 12-µm thick polyester. Before
132	operation, the detonation tube, observation chamber, and dump tank were well evacuated by an oil-sealed rotary
133	pump. Once the apparatus was evacuated, the detonation tube and observation chamber were filled with a
134	fuel-oxidizer mixture, prepared by a mixing tank. A spark plug installed at the bottom end of the detonation tube
135	was used to ignite the mixture.
136	The schlieren optical system consisted of a low-coherence, diode-laser light source (Cavitar/CAVILUX), a
137	high-speed charge-coupled device (CCD) video camera (Shimadzu/HPV-1), and some optical elements. A 150-mm
138	diameter convex lens with a 450-mm focal length was used to collimate the light emitted from the source. The
139	parallel rays of light were exposed to the observation chamber, and condensed again by a 200-mm diameter
140	concave mirror with a focal length of 1500 mm. A knife edge was positioned horizontally at the focus of the
141	concave mirror. Flashes of the light source were synchronized with exposures of the camera. The camera was
142	triggered by a pulse generator based on the signal of a pressure sensor installed in the observation chamber. The
143	emission time of the light source was set to 30 ns, and the camera exposure time was set to 250 ns. As described

- 144 below, typical propagation speed of the detonation waves  $D_{CJ}$  was approximately 2000 m/s for the conditions in
- 145 this study; the blur of the detonation front in an image could be suppressed down to 60 µm due to the short emission
- 146 time of the light source. The influence of self-luminescence was also suppressed due to the short exposure time. The
- 147 frame interval was set to 1 or 2 µs, depending on the mixture, and typical displacement of the detonation fronts between
- 148 frames could be estimated as 2 or 4 mm for  $D_{CJ} = 2000$  m/s. The spatial resolution of the obtained images was
- approximately 0.3 mm.
- 150

### 151 **2.2. Experimental conditions**

152 Experimental conditions and typical mixture characteristics are listed in Table 1. In the present study, ethylene 153 or acetylene gas, oxygen gas, and argon gas were employed for fuel, oxidizer, and diluent, respectively. As shown 154 in Table 1, four types of stoichiometric ethylene–oxygen mixtures (E1–E4) were prepared with a variation of argon 155 mole fraction  $y_{Ar}$ , as well as two types of stoichiometric acetylene-oxygen mixtures (A1-A2). For the 156 ethylene-oxygen mixture without argon dilution (E1), the inlet-channel width  $l_c$  of the observation chamber was 157 varied. Initial pressure (filling pressure) of the mixtures  $p_0$  ranged from 5–140 kPa, and initial temperature  $T_0$  was 158 at room temperature ( $22\pm6$  °C). In the present study, we focused particularly on two parameters that characterize 159 mixtures. The first was the characteristic length of detonation, and the second was detonation stability. The 160 characteristic length of detonation was varied by controlling the initial pressure of the mixture; detonation stability

- 161 was varied by changing the argon mole fraction of the mixture.
- 162 To clarify the characteristics of the mixtures, Chapman-Jouguet (CJ) detonation velocities and stability 163 parameters were calculated for the seven mixtures for a representative initial condition such that  $p_0 = 50$  kPa and  $T_0$ 164 = 22°C, as shown in Table 1. The CJ velocity was calculated using the NASA CEA code [31, 32]. As shown in 165 Table 1, CJ detonation velocity  $D_{CJ}$  decreases with an increase in the argon mole fraction  $y_{Ar}$  for the ethylene and 166 acetylene mixtures. This is the result of the lowering of exothermicity, which drives detonation. That is, in a more 167 highly diluted mixture, heat release per unit mass of the mixture decreases because of the decrease in the mass 168 fraction of the fuel. Additionally, as shown in Table 1, comparing the E1 and A1 mixtures and E4 and A2 mixtures, 169 it is evident that differences in CJ velocity due to the difference in fuel species is minor for similar argon mole 170 fractions.

171 Although some parameters have been proposed to parameterize the detonation stability of the mixture, in the 172 present study, the  $\chi$  parameter computed as follows was used as a stability parameter in accordance with reference 173 [33].

$$\chi = \frac{\tau_{\rm i}}{\tau_{\rm r}} \frac{E_{\rm a}}{RT_{\rm vN}},\tag{1}$$

where  $\tau_i$  refers to induction length,  $\tau_r$  is reaction length,  $E_a$  is overall activation energy, *R* is the gas constant, and  $T_{vN}$  is the von Neumann pressure. The stability parameter was evaluated based on a solution of the Zel'dovich-von Neumann-Döring (ZND) model simulation [34] with the GRI-Mech 3.0 chemical reaction model [35]. The

- 177 induction time was computed as the arrival time interval of the leading shock and the maximum-thermicity point.
- 178 The reaction time was computed as the inverse of the maximum thermicity. The activation energy was computed in
- accordance with the method described in Reference [23]. As shown in Table 1, with an increase in the argon mole
- 180 fraction, the stability parameter decreased from the order of  $10^1$  and approached the order of  $10^0$  for the ethylene
- 181 and acetylene mixtures. Note that in the case of the E4 mixture, a highly regular cell pattern was obtained in
- 182 Reference [36].

For each mixture, a correlation equation between the initial pressure and the cell width was derived in reference to existing experimental data, e.g., in the CALTECH detonation database [37]. The equations are as follows;

$$\lambda_{\rm E1} = 72.87 \times p_0^{-1.136}$$
 for the E1 mixture; (2)

$$\lambda_{\rm E2} = 97.96 \times p_0^{-1.140}$$
 for the E2 mixture; (3)

$$\lambda_{E3} = \sum_{j \in \{E1, E2, E4\}} \lambda_j \prod_{\substack{k \in \{E1, E2, E4\}\\k \neq j}} \frac{y_{Ar,E3} - y_{Ar,k}}{y_{Ar,j} - y_{Ar,k}}$$
for the E3 mixture; (4)

$$\lambda_{\rm E4} = 160.9 \times p_0^{-1.066}$$
 for the E4 mixture; (5)

$$\lambda_{A1} = 23.64 \times p_0^{-1.033}$$
 for the A1 mixture; (6)

$$\lambda_{A2} = 386.3 \times p_0^{-1.244}$$
 for the A2 mixture, (7)

186 where  $\lambda_{E1}$  to  $\lambda_{A2}$  are in mm and  $p_0$  is in kPa. Except for the E3 mixture, the equations were derived directly by

187 performing the power-law best fit. For the E3 mixture, the equation was developed by Lagrange interpolation from

188 the equations for the E1, E2, and E4 mixtures because experimental data were not available. The equation number

and detailed references corresponding to each mixture are listed also in Table 1.

190

## 191**3. Results and Discussion**

### 192 **3.1. Diffraction process**

193 Figure 2 present representative results of the sequential schlieren imaging, supercritical and subcritical 194 detonation processes for the stoichiometric ethylene-oxygen mixture without argon dilution (E1), and a channel 195 width  $l_c$  of 10 mm, with a frame interval of 2  $\mu$ s. The order of the photographs corresponds to the time evolution of 196 the processes. Note that the sequences of schlieren photographs were obtained for single diffraction events. 197 The supercritical diffraction process shown in Fig. 2 (a) (and Movie 1a, available as supplementary material) 198 was obtained for the conditions of the mixture where initial pressure and temperature were 30 kPa and 20 °C, 199 respectively. Note that the images were originally obtained with a frame interval of 1  $\mu$ s, but that some were 200 omitted because of space limitations. In Fig. 2 (a-1), a planar detonation wave generated in the detonation tube can 201 be observed in the inlet channel. The detonation wave then reached the end of the inlet channel and experienced the 202 abrupt expansion of the channel between Figs. 2 (a-1) and (a-2). After emerging into wider space (Figs. 2 (a-1) – 203 (a-5)), the detonation wave front underwent deformation gradually, due to the expansion waves generated at the end 204 corner of the inlet channel. Particularly in the vicinity of the horizontal wall, the reaction front was decoupled from 205 the shock front, whereas coupling of the shock front and reaction front was retained in the vicinity of the vertical

206	wall, as shown in Figs. 2 (a-2) – (a-9). This behavior has been interpreted as transverse waves accompanying the
207	detonation front being more significantly attenuated in the vicinity of the horizontal wall by the expansion waves
208	generated at the end corner of the inlet channel. In Fig. 2 (a-6), a bulge can be observed at the upper-right part of
209	the detonation front, which is an indication of a local explosion, i.e., re-initiation of the transverse detonation. In
210	Figs. 2 (a-6) – (a-9), the bulge evolved into a transverse detonation wave that swept the shocked but unreacted
211	region, i.e., the region between the decoupled shock and reaction fronts, toward the horizontal wall. Finally, the
212	transverse detonation wave reached the horizontal wall, as shown in Fig. 2 (a-10). At this point, the detonation
213	wave front that had detached from the horizontal wall temporarily upon emergence into the wider space reattached
214	to that wall. Later, in Figs. 2 (a-11) and (a-12), the transverse detonation wave was reflected on the wall.
215	As mentioned in the Introduction, it is known that the locus of reflection of the transverse detonation on the
216	horizontal wall behaves in an orderly way [2, 23, 28]. Thus, in the present study, we redefine the distance between
217	the reflection point of the transverse detonation wave and the end corner of the inlet channel as a novel
218	characteristic length of detonation phenomena, and we refer to it as the <i>reflection point distance</i> $l_{\rm r}$ . How to
219	determine the locus of reflection of the transverse detonation is examined in more depth below, in conjunction with
220	the reflection event.
221	Figure 2 (c) shows schematic illustrations of the temporal development of a reflection event in a process of
222	supercritical diffraction. The illustrations were rendered based on the schlieren images obtained for the same

223	operation shown in Fig. 2 (a). In the process of supercritical diffraction, the point of intersection between the
224	transverse detonation wave and the decoupled shock front arose after a local explosion, as shown in Figs. 2 (a-8)
225	and Fig. 2 (c-1). The point of intersection moved along the decoupled shock front with the propagation of the
226	transverse detonation until finally reaching the horizontal wall, as shown in Figs. 2 (a-9) and (a-10). Thus, a
227	reflection point distance $l_r$ was quantified based on a high-speed schlieren image as the distance from the exit
228	corner of the channel to the locus where the intersection arrives at the horizontal wall, as shown in Fig. 2 (c-2).
229	Here, the pressure distribution is thought to have a locally maximum value at the point of intersection between the
230	transverse detonation and decoupled shock front. Thus, the reflection point distance defined above corresponds to
231	the circular patterns in the soot foil records on the exit face described in [28], which can be interpreted as the
232	history of the locus of locally highest pressure. In the following subsection, we consider the characteristics of the
233	reflection point distance.
234	The subcritical diffraction process shown in Fig 2 (b) (and Movie 1b, available as supplementary material)
235	was obtained for the conditions of the mixture where the initial pressure and temperature were 10 kPa and 20 °C,
236	respectively. In Fig. 2 (b-1), as in the supercritical case, a planar detonation wave can be observed in the inlet
237	channel. Between Figs. 2 (b-1) and (b-2), the detonation wave emerged into wider space. The expansion wave was
238	then generated at the end corner of the inlet channel, and similarly propagated leftward while the detonation wave
239	front underwent deformation, as shown in Figs. 2 (b-2) – (b-6). Over the course of these figures, in contrast to the

- supercritical case, the expansion waves attenuated the transverse waves accompanying the detonation front more significantly, and the reaction front was immediately decoupled from the shock front upon the passing of the head of the expansion waves.
- 243 By the time of Fig. 2 (b-7) or (b-8), the head of the expansion fan reached the vertical wall, and the reaction
- front totally decoupled from the shock front. The quenching mechanism in this case can be classified into the local
- 245 quenching mode described by Lee [1]. After Fig. 2 (b-8), the shock front was decelerated, which can be observed
- by comparing Fig. 2 (b) to 2 (a). That is, from Figs. 2 (b-1) to (b-6) or (b-7), the locus of the detonation front on the
- 247 vertical wall coincides with the one observed in Figs. 2 (a-1) to (a-6) or (a-7), whereas after Fig. 2 (b-8), the
- 248 difference in the loci of the detonation front becomes evident. Additionally, in Figs. (b-4) (b-12), attenuated
- transverse wave remnants can be observed, and the spacing between them expands with time.

250

### 251 **3.2. Reflection point distance**

Before entering into the details of the results , the nature of the reflection point distance is briefly discussed here. The reflection point distance should be essentially defined in a system with an infinitely wide donor channel. In such an ideal system, there is no length scale characterizing the geometry, and thus the features of the flow-field are determined based only on physical and chemical properties intrinsic to the mixture [49]. That is, in such a system, the reflection point distance can be a characteristic length manifesting a property of the detonable mixture.

257	In the present experiments, in reality, the channel width must be, of course, finite, and thus the wall of the channel
258	affects the flow-field by reflecting transverse waves of the detonation and the rarefaction wave generated at the
259	corner of the channel. However, by using a wide enough donor channel compared to the transverse wave spacing of
260	the detonation wave, influence of the physical boundary is expected to be suppressed. Hence, at high initial
261	pressures where the transverse wave spacing is small, the reflection point distances obtained in experiments are
262	substantially equivalent to the ideal ones, aside from the inherent variation arising from the stochastic nature of the
263	detonation diffraction problem.
264	In Figs. 3 and 4, as a summary of data reduction results from the schlieren images, reflection point distance $l_r$
265	as a function of the initial pressure $p_0$ of the mixture is shown in a double-log scale for the stoichiometric
266	ethylene-oxygen mixtures (E1–E4) with different argon mole fraction $y_{Ar}$ , as well as the stoichiometric
267	acetylene-oxygen mixtures (A1-A2). The circles correspond to the ethylene mixtures (E1-E4), and the squares to
268	the acetylene mixtures (A1–A2). The sizes of the symbols correspond to the argon mole fraction $y_{Ar}$ , i.e., the
269	smaller the symbol, the more highly diluted the mixture. The closed symbols refer to the case where channel width
270	$l_c = 10$ mm, whereas the open symbols indicate the case where channel width $l_c = 20$ mm. The dashed auxiliary
271	lines indicate inversely proportional relations as an aid in visualization.
272	Based only on Fig. 3, we describe the influence of the initial pressure $p_0$ , argon mole fraction $y_{Ar}$ , and channel
273	width $l_c$ , focusing on the ethylene mixtures (E1-E4). For constant argon mole fractions, the reflection point

274	distance $l_r$ decreases with the increase in the initial pressure of the mixture. The relations between the reflection
275	point distance and the initial pressure appear to be roughly inversely proportional, although slight deviations can be
276	seen at low pressures (e.g., $y_{Ar} = 0$ %, $l_c = 20$ mm, and $p_0 = 15$ kPa). This deviation can be attributed to the fact that
277	the channel width was not sufficiently wider than the transverse wave spacing at those initial pressures. For
278	constant initial pressures, the reflection point distance increases with the argon mole fraction; e.g., in Fig. 3, at $l_c$ =
279	10 mm and $p_0 = 50$ kPa, $l_r = 11.2$ , 23.9, and 41.2 mm for $y_{Ar} = 0$ %, 50%, and 67%, respectively. The argon mole
280	fraction also seems to affect the scattering of the reflection point distance. Namely, the scattering of the reflection
281	point distance becomes more significant as the argon mole fraction increases. Additionally, for the E1 mixture ( $y_{Ar}$
282	= 0 %), the reflection point distance appears to be not significantly influenced by the change in the channel width,
283	and reflection point distances are similar for both the channel widths ( $l_c = 10$ and 20 mm), particularly at high
284	initial pressures. This fact suggests that the channel is wide enough, and reflection point distances obtained at such
285	high initial pressures are close to the ideal ones.
286	By comparing Figs. 3 and 4, the influence of the fuel species can also be deduced. First, descriptions of the
287	influences of the initial pressure and argon mole fraction for the ethylene mixtures (E1-E4) above are also
288	applicable to the acetylene mixtures (A1-A2). Second, for identical initial pressure, argon mole fraction, and
289	channel width, the reflection point distance is smaller for the non-diluted stoichiometric acetylene-oxygen mixture
290	(A1) than for the non-diluted stoichiometric ethylene-oxygen mixture (E1), e.g., at $p_0 = 50$ kPa, $y_{Ar} = 0\%$ , and $l_c = 10\%$



292 As another quantity relevant to supercritical diffraction, re-initiation distance has been also discussed, e.g., by 293 Edwards et al. [8], Nagura et al. [30], and Gallier et al. [50]. When a re-initiation distance is defined as a distance 294 between a re-initiation point and the channel end face, the re-initiation distance was approximately 60-90% of the 295 reflection point distance in this study. Additionally, the behavior of the re-initiation distance was similar to the 296 behavior of the reflection point distance. Namely, the re-initiation distance was again roughly inversely 297 proportional to the initial pressure of the mixture and independent of the channel width; with diluent (argon) mole 298 fraction, the re-initiation distance increased and its scattering was enhanced; the re-initiation distance was smaller 299 for the non-diluted acetylene mixture A1 than for the non-diluted ethylene mixture E1 under an identical initial 300 pressure, an argon mole fraction, and a channel width. It is, however, not easy to define the re-initiation distance in 301 an objective fashion depending on a clear criterion because the very onset of the re-initiation is difficult to identify 302 by the method employed in this study. Thus, the following discussion focuses only on the results for the reflection 303 distance.

In Figs. 5 and 6, the reflection point distance  $l_r$  as a function of the initial partial pressure of the fuel  $p_{0,f}$  for the ethylene mixtures (E1–E4) and acetylene mixtures (A1–A2), respectively, are shown in a double log-scale. The circles correspond to the ethylene mixtures (E1–E4), and the squares to the acetylene mixtures (A1–A2). A smaller symbol corresponds to a more highly argon-diluted mixture. The closed and open symbols refer to the cases where

308	channel width $l_c = 10$ and 20 mm, respectively. In the figures, dashed lines represent the best-fit curves obtained by
309	assuming that the reflection point distance $l_r$ is inversely proportional to the initial partial pressure of the fuel $p_{0,f}$ .
310	The fitting parameters were the proportionality constants, i.e., the product of the reflection point distance and initial
311	partial pressure of the fuel, $l_r p_{0,f}$ . Note that the fitting was performed only for the results which satisfy $l_r/l_c < 4$
312	because, as mentioned above, the reflection point distance obtained in the experiment is substantially equivalent to
313	the ideal value at high enough initial pressures. Judging from the results shown in Figs. 3 and 4, this criterion seems
314	plausible, albeit somewhat arbitrary. The determined proportionality constants are also shown in the figures.
315	Additionally, in Fig. 7, the determined proportionality constants vs. the argon mole fractions are summarized for
316	the ethylene and acetylene mixtures. The circles correspond to the ethylene mixtures (E1-E4), and the squares to
317	the acetylene mixtures (A1-A2). A smaller symbol corresponds to a more highly argon-diluted mixture. Note that
318	only one fitting curve is provided for the E1 mixture in Fig. 5, although there is a variation in the channel width,
319	because the influence of the channel width on the reflection point distance is minor, as described above.
320	As can be seen in Figs. 5 and 6, for identical combinations of fuel and oxidizer, the relationship between
321	reflection point distance $l_r$ and initial partial pressure of fuel $p_{0,f}$ is roughly identical, although the reflection point
322	distance tends to increase slightly with the argon mole fraction for the same initial partial fuel pressure. Here, it is
323	also evident that the fitted curves with function forms where reflection point distance is inversely proportional to
324	initial partial pressure of fuel correspond well to the relationship between reflection point distance $l_r$ and initial

325	partial fuel pressure $p_{0,f}$ . As shown in Fig. 7, the proportionality constant slightly increases with the argon mole
326	fraction. Additionally, the constant is smaller for acetylene mixtures than for the ethylene mixtures. The rough
327	identity of the reflection point distance as a function of the initial partial pressure of fuel among different argon
328	mole fractions means that the reflection point distance is determined mainly by the amount of heat release per
329	volume. The slight differences with variation in argon mole fraction are thought to be due to slight changes in the
330	thermodynamic and chemical properties of the mixtures, including heat capacity, specific heat ratio, and reaction
331	heat, with a change in the argon mole fraction.
332	In Figs. 8 and 9, the ratios of <i>re-evaluated</i> reflection point distance to the channel width, $l_r/l_c$ , are plotted
333	against the argon mole fraction $y_{Ar}$ for the ethylene mixtures (E1–E4) and acetylene mixtures (A1–A2), respectively
334	The circles and squares correspond to supercritical diffraction in the ethylene and acetylene mixtures, respectively.
335	The closed and open symbols refer to the cases where channel width $l_c = 10$ and 20 mm, respectively. The
336	reflection point distance $l_r$ was re-evaluated by using the fitted curves shown in Figs. 5 and 6, which represent
337	functional relationships between the reflection point distance and the initial partial pressure of fuel. That is, for a
338	given initial partial pressure of fuel, each data point is given through a calculation using the corresponding fitted
339	curve. Note that data points were given also for the subcritical cases at relatively low initial pressures, where
340	re-initiation does not occur in reality, by extrapolation from data at high initial pressures, because ideally, the
341	reflection point distance is suggested to be roughly inversely proportional to the initial pressure as described above.

0.40					
342	In Figs 8 and 9	the data noints extr	anolated for subcritical	cases are indicated h	$v \times$ -mark symbols
012	$111165.0$ and $\gamma$ ,	ine data points extr	applated for subernieur	cuses are maleated b	y ··· mark symbols.

343 When the channel width is decreased gradually under certain mixture conditions, detonation transmission from 344 the channel to unconfined space will fail at some point. Then, that channel width is the critical width for those 345 mixture conditions. Here, by analogy from the cell width, the critical channel width is expected to be generalized 346 by making it a dimensionless quantity with the use of the ideal reflection point distance, which can serve as a 347 characteristic length of the mixture as described above. Thus, in this study, critical condition is given by the ratio of 348 ideal reflection point distance to the channel width,  $l_{\rm r}/l_{\rm c}$ . Note that the reflection point distance has relatively large 349 values as a characteristic length, and thus the ratio of the reflection point distance to the channel width is more 350 convenient to express the critical condition rather than its inverse, although the critical condition is usually given by 351 dividing the channel size by a characteristic length of detonation such as the cell width. 352 If we assume that the critical conditions  $(l_r/l_c)_{crit}$  lie in a range of 3–5, which is shown by a shaded band in Figs. 353 8 and 9, for all the ethylene and acetylene mixtures with different argon mole fractions, the results shown in Figs. 8 354 and 9 are consistent with the assumption because the critical value of the ratio  $l_r/l_c$  is thought to exist between the 355 largest value of the ratio  $l_r/l_c$  in the supercritical cases and the smallest value of the ratio  $l_r/l_c$  in the subcritical cases 356 for each mixture, whether the outcome overlaps or not. As shown in Table 1, the stability of the mixture increases 357 with the argon mole fraction, thus the critical condition may also vary  $(l_r/l_c)_{crit}$  with the argon mole fraction. 358 However, the dependence of the critical condition on the stability cannot be deduced clearly only from the results

- 359 obtained in this study, although the dependence is potentially small.
- 360 In Figs. 10 and 11, the ratios of the channel width to the cell width,  $l_c/\lambda$ , are plotted against the argon mole 361 fraction  $y_{Ar}$  for the ethylene mixtures (E1–E4) as well as the acetylene mixtures (A1–A2), respectively. The closed 362 and open symbols refer to the cases where channel width  $l_c = 10$  and 20 mm, respectively. The ×-mark symbols 363 correspond to subcritical diffraction for all the mixtures. As can be seen in Figs. 10 and 11, the critical value for the 364 ratio  $l_c/\lambda$ , which is thought to exist between the smallest value of the ratio  $l_c/\lambda$  in supercritical diffraction and the 365 largest value of the ratio  $l_c/\lambda$  in subcritical diffraction for each mixture, is not universal but dependent on the argon 366 mole fraction, i.e., stability, as has been well known (e.g. [16, 19]). The critical value  $(l_c/\lambda)_{crit}$  appears to be greater 367 for a more highly argon diluted mixture. Here, in the cylindrical (two-dimensional) diffraction problem, the critical 368 value of  $(l_c/\lambda)_{crit} \sim 1.5$  has been given, e.g., by Liu *et al.* [11], Benedick *et al.* [12], and Thomas et al. [13] for more 369 unstable mixtures like E1 and A1 in this study, and the critical value  $(l_c/\lambda)_{crit} \sim 6$  has also been given by Meredith et 370 al. [17] for a mixture with high argon dilution featuring a more regular cellular structure. Additionally, Shepherd et 371 al. [14] suggested that for stoichiometric acetylene-oxygen mixtures, the critical value  $(l_c/\lambda)_{crit}$  was doubled by the 372 increase in the argon mole fraction from 0% (corresponding to the A1 mixture) to 80% (corresponding to the A2 373 mixture), although the critical values were obtained in the spherical (three-dimensional) diffraction problem. Hence, 374 as can be confirmed from Figs. 10 and 11, the results obtained in this study are consistent with known correlations. 375 Note that the flow-fields obtained in the present setup should be regarded as right halves of symmetric flow-fields

376	in cylindrical diffraction, and the vertical wall of the observation chamber corresponds to the line of symmetry, thus
377	the critical ratios in this study were also halved. It can be also important to point out the difference in the
378	dependence of the critical condition on the argon mole fraction, i.e., stability. Namely, for the scale of the cell width,
379	the critical condition is significantly dependent on the argon mole fraction, and the critical value at $y_{Ar} = 75\%$ is
380	about four times the value at $y_{Ar} = 0\%$ as shown in Fig. 10, whereas for the scale of the reflection point distance, the
381	critical value at $y_{Ar} = 0\%$ is at most five-thirds times the value at $y_{Ar} = 75\%$ as shown in Fig. 8. That is, the critical
382	condition given by the reflection point distance may be less sensitive to the stability of the mixture. This suggests
383	that the diffraction problem is not substantially governed by the cellular spacing.
384	Finally, significant features of the reflection point distance as a characteristic length are elaborated here
385	through comparison with the cell width. As confirmed in previous studies [29, 30, 36], the reflection point distance
386	was roughly proportional to the cell width and was tens of times the cell width, i.e., the reflection point distance has
387	significantly greater values compared to the cell width. This is particularly important in determining the
388	characteristic lengths of detonable mixtures at relatively high initial pressures, which are encountered in practical
389	systems, including pulse and rotating detonation engines. For instance, for non-diluted fuel-oxygen mixtures at
390	initial pressures greater than the atmospheric pressure, cell widths can be often less than millimeters. Additionally,
391	by using special equipment-such as the apparatus in this study-the reflection point distance may be less
392	cumbersome to determine than the cell width, owing to avoidance of careful preparation of soot foil and

393	problematic interpretation of the pattern recorded on the foil. It should be also noted that the reflection point
394	distance reflects effects of the three-dimensional cellular structure of the detonation wave because the reflection
395	distance strongly depends on timing of the re-initiation, which again depends on the cellular structure. Here, the
396	usefulness of the cell width is evident in interpreting detonation phenomena, and there exists a huge collection of
397	data. Nevertheless, introduction of the reflection point distance as a characteristic length can still potentially
398	provide a novel point of view. A much wider range of experiments to verify the generality of the reflection point
399	distance parameter is future work.
400	
401	4. Conclusions
402	For stoichiometric C <sub>2</sub> H <sub>4</sub> –O <sub>2</sub> and C <sub>2</sub> H <sub>2</sub> –O <sub>2</sub> mixtures with or without argon dilution, the processes of detonation
403	diffraction have been investigated in a two-dimensional setup through high-speed schlieren imaging with variations
404	of characteristic length and detonation stability by regulating the initial pressure and argon mole fraction of the
405	mixture. In supercritical cases, reflection point distances of the transverse detonation on the end face of the donor
406	channel were particularly deduced from the obtained sequential schlieren images and analyzed. The reflection point
407	distance can be idealized for the infinitely wide donor channel because of the absence of a length scale
408	characterizing the geometric setup of this problem, and thus it can be a parameter in which properties intrinsic to
409	each detonable mixture are manifested. We draw the following conclusions from our results.
410	The reflection point distance was roughly inversely proportional to the initial pressure for identical mixtures

- 411 and independent of the width of the donor channel at high initial pressures. For a certain combination of the fuel
- 412 and oxidizer, correlations between the reflection point distance and the initial partial pressure of fuel were very
- 413 similar regardless of the argon mole fraction. Critical conditions of the diffraction problem could be given for the
- 414 ratio of the reflection point distance to the channel width, and it was suggested that the critical value lies in a range
- 415 of 3–5 and does not significantly depend on the stability of the mixture.
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Table 1 Experimental conditions and typical mixture characteristics. CJ detonation velocities and stability parameters were calculated using the NASA CEA code [31, 32] and ZND code [34] with GRI-Mech 3.0 [35], respectively, at  $p_0 = 50$  kPa and  $T_0 = 293$  K.

			, [35], iespe	centrely, at p	-0 50 M	a ana 10	2)) IX.
	Mixture	УАг [%]	l <sub>c</sub> [mm]	D <sub>CJ</sub> [m/s]	χ	λ	Cell data
E1	$C_2H_4+3O_2/Ar$	0	10, 20	2340	26	Eq. 2	[10, 38, 39]
E2		50	10	1940	9.7	Eq. 3	[38, 40]
E3		67	10	1800	6.3	Eq. 4	
E4		75	10	1720	4.9	Eq. 5	[38]
A1	$2C_{2}H_{2} + 5O_{2}/Ar$	0	10	2390	28	Eq. 6	[16, 41–48]
A2		80	10	1690	5.9	Eq. 7	[16, 48]



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Fig. 2. Typical sequences of single detonation diffraction processes for  $C_2H_4 + 3O_2$  (E1) and  $l_c = 10$  mm. (a) Schlieren photographs for supercritical diffraction,  $p_0 = 30$  kPa,  $T_0 = 20$  °C, frame interval = 2 µs. See also Movie 1a, available as supplementary material. (b) Schlieren photographs for subcritical diffraction,  $p_0 = 10$  kPa,  $T_0 =$ 20 °C, frame interval = 2 µs. See also Movie 1b, available as supplementary material. (c) Illustration of a reflection event of a transverse detonation wave in supercritical diffraction, frame interval = 1 µs.

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80 70 60 50 67% 75% 50% 0% = **y**<sub>Ar</sub> The second se 0 40 30 20 /<sub>r</sub> [mm] 10  $I_{c} = 10 \text{ mm}^{2}$ 20 mm 10<sup>1</sup> 10<sup>2</sup> *p*<sub>0</sub> [kPa]

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Fig. 3. Reflection point distance vs. initial pressure for supercritical diffraction in ethylene mixtures (E1–E4).



610 Fig. 4. Reflection point distance vs. initial pressure for supercritical diffraction in acetylene mixtures (A1–A2).

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- 70 60 75% 67% 50% **y**<sub>Ar</sub> 0% 50 40 30 /<sub>r</sub> [mm] 20 *I*<sub>r</sub>*p*<sub>0,f</sub>= 209 m Pa 165 m Pa 151 m Pa 152 m Pa 10 /<sub>c</sub> = 20 mm ≁ 10, mm 5 15 3 10 *p*<sub>0,f</sub> [kPa]
- 618

619 Fig. 5. Reflection point distance vs. initial partial pressure of fuel for supercritical diffraction in ethylene mixtures620 (E1-E4).



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Fig. 6. Reflection point distance vs. initial partial pressure of fuel for supercritical diffraction in acetylene mixtures(A1-A2).

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Fig. 7. Product of reflection point distance and initial pressure vs. argon mole fraction for supercritical diffraction.

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8 = 152 m P  $\boldsymbol{p}_{0,f}$ 7 XX 6 Х **P**<sub>0,f</sub> subcritical Х 5 X X 4 \_\_\_\_ 0 3 0 0 0 0 0 0 supercritical • 209 m Pa 2 0 0000 165 m Pa = 10 mm 1 = 20 mm 151 m Pa 0 40 60 0 20 80 100 **y<sub>Ar</sub> [%]** 

Fig. 8. Ratio of re-evaluated reflection point distance to channel width vs. argon mole fraction in ethylene mixtures
(E1-E4). The reflection point distances were re-evaluated by using the fitted curves shown in Fig. 5. For the
subcritical diffraction, data points were given by extrapolation.

- 649
- 650

- - -



Fig. 9. Ratio of re-evaluated reflection point distance to channel width vs. argon mole fraction in acetylene
mixtures (A1–A2). The reflection point distances were re-evaluated by using the fitted curves shown in Fig. 6. For
the subcritical diffraction, data points were given by extrapolation.

20 mm **I**, / λ 10 mm percritical **p**<sub>0,f</sub> critical 40 6 *Y*<sub>Ar</sub> [%] 

Fig. 10. Ratio of channel width to cell width vs. argon mole fraction in ethylene mixtures (E1–E4).



Fig. 11. Ratio of channel width to cell width vs. argon mole fraction in acetylene mixtures (A1–A2).