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1	Experimental study on evaporation characteristics of
2	light cycle oil (LCO) droplet under various ambient
3	conditions
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14	KEYWORDS: Droplet evaporation; Low ignitability fuel; Cetane index; Low sulfur fuel; Marine
15	fuel

16 ABSTRACT: The authors conducted droplet evaporation experiments of light cycle oil (LCO) at 17 various ambient temperatures and pressures. Five kinds of LCO and three kinds of arranged-fuels 18 were used. We investigated the evaporation characteristics of LCO and the relationships between 19 the evaporation characteristics and the cetane index. In addition to that, a surrogate fuel composed 20 of 4 chemical species, which can simulate the droplet evaporation characteristics of LCO, was 21 suggested. Experimental results show that the differences in droplet lifetime between fuel species 22 become larger with decreasing ambient temperature. This is because the low volatile component 23 made the evaporation rate outstandingly slow at a low ambient temperature. Second, it was found 24 that the relationship between droplet lifetime and the late-stage distillation temperature becomes 25 stronger at low ambient temperature and high ambient pressure. By an analysis employing the 26 properties of chemical species in LCO surrogate fuel, it is clarified that the mass evaporation rate becomes smaller than the internal diffusion, which is the condition similar to that in the distillation 27 28 test. Finally, the relationship between the droplet lifetime and the cetane index was investigated. 29 It can be concluded that the droplet lifetime is independent of the cetane index under all conditions 30 tested in this study. The experimental data obtained by this research can be utilized for the 31 validation of multi-component fuel droplet evaporation models in the future.

32

33 1. INTRODUCTION

34 Spray combustion is widely employed in industrial applications such as internal combustion engines, gas turbine engines, and rocket propulsion systems. However, the unstable combustion 35 phenomena which cause a malfunction of the combustor has been reported.^{1,2} For instance, 36 37 knocking has been recognized as one of major problems for the reciprocating engine. An ignition 38 delay of fuel is an important factor determining the occurrence of the knocking. To investigate the 39 relationship between the ignition delay and the knocking, a study for single droplet ignition was 40 conducted by some reserachers.³ Furthermore, the ignition delay and combustion characteristics 41 of biodiesel also have been studied because the practical usage of the next-generation fuel for an internal combustion engine is attempted in recent years.^{4,5} Even though relationship between the 42 43 combustion characteristics and the ignition delay has been investigated a lot, dominant factors for 44 the ignition delay has not been clarified because the structure of spray combustion is much 45 complicated, which is including atomization, droplet evaporation, and chemical reactions. 46 Therefore, fundamental knowledge to clarify the dominant factors in the ignition delay is required. 47 To clarify the complex mechanism of spray combustion, a numerical simulation tool has been 48 strongly expanded. By employing the numerical simulation, not only the characteristics of spray⁶ and spray flame such as jet flame^{7,8} but also elements of spray flame such as atomization⁹ and 49 impingement¹⁰ have been investigated. In particular, the prediction of fuel droplet size 50 51 distributions and evaporation rate could affect the prediction of combustion properties including efficiency, stability, and pollutant formation rate. Noh et. al.¹¹ compared the spray combustion 52 properties predicted by the different types of droplet evaporation models. As one of their findings, 53 the predicted flame lift-off height depends on the droplet evaporation rate. It was revealed that the 54 55 accuracy of a droplet evaporation model strongly influences the accuracy of the spray flame

structure predictions. Hence, the droplet evaporation model is one of the most important parts of the numerical simulation technology for the prediction of the spray combustion fields of the combustor. To develop the accurate droplet evaporation model, the experimental data for droplet evaporation is necessary for its validation. Nomura *et al.* investigated the droplet effect of ambient pressure on the evaporation characteristics of n-heptane¹² and n-hexadecane¹³.

The droplet evaporation experimental data for market fuels have been also obtained by some 61 researchers, while droplet combustion which is single droplet^{14,15} as well as droplet cloud¹⁶ have 62 been studied a lot. For example, Elkotb et al.¹⁷ studied the evaporation of commercial fuels (i.e., 63 64 heavy diesel fuel, light diesel fuel, kerosene, gasoline, and their blends) at 400 °C and atmospheric pressure. Morin *et al.*¹⁸ investigated the vaporization of vegetable oil droplets used as biofuels at 65 high ambient temperature. Ghassemi et al.¹⁹ investigated the vaporization of kerosene droplet at 66 high temperatures and high pressures. Hashimoto et al.²⁰ investigated the evaporation 67 68 characteristics of a palm methyl ester droplet at high ambient temperature. However, the 69 experimental data of multi-component market fuel droplet evaporation are still very limited. For 70 instance, the experimental data of droplet evaporation for light cycle oil (LCO) have not been 71 obtained. The following is the reason why the experimental data of droplet evaporation for LCO 72 is required.

As one of the significant environmental regulations, the International Maritime Organization (IMO) decided to tighten the global regulations of sulfur content in maritime fuel used on-board ships from 3.5% to 0.5% since 2020. To reduce the sulfur oxides in the exhaust gas, the fuel with low sulfur contents could lead to finding a solution to the root cause. Therefore, the efficient use of cracked middle distillates that contain fewer sulfur compositions is becoming important because of the increasing demand for diesel fuel.²¹ As one of the cracked middle distillates, light cycle oil (LCO) is a promising fuel of commercial diesel fuel, which is already mixed in maritime fuel as the fuel oil base material.²² In response to the IMO regulations, some researchers investigated the fuel properties of LCO for practical usage and market growth.^{23,24}. Hence, to correspond to the high market demand for LCO, the effect of using LCO on a combustor should be investigated.

Despite the advantages of LCO such as low sulfur compositions, LCO tend to contain a lot of 83 84 aromatic compositions and be characterized as low ignitability fuel. Therefore, considering the 85 increase of the situation that LCO is used as a part of a mixture product, the influence of using LCO on the diesel engines should be clarified. Xu et al.^{25,26} investigated the combustion of LCO 86 single droplets in normal and microgravity conditions. Imhof et al. 27 investigated emissions of 87 LCO water-in-fuel emulsions with a test engine to reduce NOx and unburned emission to follow 88 IMO regulation. Tashima et. al.²⁸ demonstrated computational fluid dynamics (CFD) simulation 89 90 including spray development and combustion process by using the fuel blending LCO. 91 Furthermore, to develop the high precision simulation technique for LCO spray combustion, Takagi et. al.²⁹ developed a droplet evaporation model for multi-component fuel to investigate the 92 93 relationships between evaporation characteristics and ignition delay. They numerically clarified 94 that the effect of evaporation time on ignition delay become larger with increasing cetane index. 95 Furthermore, they compared the predicted droplet evaporation characteristics of 13 kinds of LCO 96 to clarify the relationship between the ignitability index and evaporation characteristics. However, 97 the accuracy of the droplet evaporation model for LCO has not been clarified yet because the 98 experimental data of LCO droplet evaporation have not been reported yet. To develop precise 99 numerical simulation technologies for LCO spray combustion, the accuracy of the droplet 100 evaporation model, which can affect the simulation results significantly, should be verified well 101 by using experimental data.

In this study, the droplet evaporation characteristics of LCO at various ambient temperatures and pressures were investigated. The relationships between the evaporation characteristics and fuel ignitability index are investigated. The experimental data obtained by this study can be utilized for the future development of numerical simulation technology for LCO spray combustion as the validation data.

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108 2. EXPERIMENTAL METHOD

109 2.1 LCO properties

110 This study used five kinds of LCO with different properties. Table 1 shows the properties of the 111 LCO. L1, L2, L3, L12, and L13 are LCOs obtained at different oil refineries. L1, L2, L3 are the 112 combination to investigate the effect of the differences of distillation characteristics. L12 L13 is 113 the combination to have similar distillation characteristics. The fuels used in this study cover the hydrocarbon compositions of LCO not only before but also after hydrogenation.³⁰ As one of the 114 115 remarkable characteristics of LCO, its cetane index is generally smaller than that of other 116 transportation fuels, such as light oil and A-type heavy oil, which is why LCO is generally well 117 known as a low-ignitibility fuel. The cetane index is applied instead of the cetane number when 118 the cetane number cannot be obtained by the prescribed test engine. In this study, two kinds of 119 cetane index are introduced. First on is the old cetane index (OCI) based on ASTM D976-66 or 120 JIS K 2204:1983, which is expressed as follows:

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$$OCI = 0.49083 + 1.06577X - 0.0010552X^{2}$$
(1)

$$X = 97.833(\log_{10}(A))^{2} + 2.2088B_{0}\log_{10}(A) + 0.0124B_{0}^{2} - 423.51\log_{10}(A)$$
(2)

$$- 4.7808B_{0} + 419.59$$
(2)

$$A = \frac{9}{5}T_{50} + 32\tag{3}$$

122

where B_0 is the API gravity (-), and T_{50} is 50% volume distillation temperature (°C). Only 50% volume distillation temperature is considered in the OCI. Second one is the new cetane index (NCI) based on ISO 4264:2007 or JIS K 2280-5:2013, which is expressed as follows:

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$$NCI = 45.2 + 0.0892(T_{10} - 215) + (0.131 + 0.901B)(T_{50} - 260) + (0.0523 - 0.42B)(T_{90} - 310) + 0.00049\{(T_{10} - 215)^2 (4) - (T_{90} - 310)^2\} + 107B + 60B^2 B = exp\{-0.0035(1000D - 850)\} - 1$$
(5)

127

where T_{10} is the 10% volume distillation temperature (°C), T_{50} is the 50% volume distillation temperature (°C), T_{90} is the 90% volume distillation temperature (°C), and D is the density at 15 °C (g/cm³). In the NCI, 50%, 10%, and 90% volume distillation temperatures are considered, while only 50% volume distillation temperature is considered in the OCI. Note that in LCO, Takagi *et al.* ³¹ clarified that the ignition delay of LCO, which has the equivalent OCI to standard fuel of cetane number, can be related to that of the standard fuel of cetane number. However, the relationship between CI and evaporation characteristics has not been clarified.

As the other characteristics of LCO, a large number of aromatic hydrocarbons are contained in comparison with the other transportation fuels. Table1 shows that L1, L2, and L3 contains significant amounts of aromatic hydrocarbons. L1 and L3 contain over 80vol% aromatic hydrocarbons. In particular, L3 has over 20vol% aromatic hydrocarbons with at least three rings, which is the largest amount in the present LCO. L2 contains the largest amount of olefin in the

140	present LCO. L12 and L13 contain a large
141	number of saturated hydrocarbons that are
142	over 50vol%. Figure 1 shows the distillation
143	curves of LCO and light oil ²⁰ . According to
144	Figure 1, L1 has the minimum difference
145	between the initial boiling point (0%) and the
146	ending point (100%) in LCO. On the other
147	hands, L2 contains the largest number of the
148	component with a low boiling point in LCO.

Regarding the ending point of the distillation



Figure 1. Distillation curves of LCO, arranged-fuels and light oil.

curve, L3 contains components with the highest boiling point. On the contrary, L12 and L13 have
similar distillation characteristics. Note that the specific distillation temperatures in Figure 1 are
printed in Supporting Information.

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154 2.2 Arranged-fuels (S30, S35 and S40)

Arranged-fuels (S30, S35, and S40) were used to investigate the effect of the differences between the OCI and NCI on the evaporation characteristics of LCO. The arranged-fuels, are made to have almost the same NCI, are the liquid mixtures of refinery feedstocks and solvents. LCO used in this study has a variety of not only distillation characteristics but also cetane index. This makes it difficult to clarify whether the distillation characteristics or the cetane index have related to the evaporation characteristics. To clarify the relationship between the cetane index and droplet evaporation characteristics, the authors made the arranged-fuels, which has different distillation 162 characteristics but almost the same cetane index. The properties of the arranged-fuels are shown

- in Table 1.
- 164

Table 1. Properties of light cycle oil and arranged-fuel used in this study

			Light Cycle Oil				Arranged-Fuel			
			L1	L2	L3	L12	L13	S30	S35	S40
Density(15°C)		g/cm ³	0.9323	0.9266	0.9659	0.9018	0.8931	0.8834	0.8968	0.9028
Flash point		°C	68	52	N/A	N/A	67	29	79	82
Dynamic vis	cosity(30°C)	$\mathrm{mm^2/s}$	2.169	2.097	N/A	N/A	3.425	2.454	3.844	4.582
Dynamic viscosity(50°C)		$\mathrm{mm^2/s}$	1.522	1.493	2.469	2.295	2.256	1.724	2.497	2.898
Water content		wt%	0.00	0.00	N/A	N/A	0.00	0.00	0.00	0.00
Pour point		°C	<-45	<-45	N/A	N/A	-2.5	-30.0	-22.5	-22.5
CC	CAI	-	911	907	935	875	855	857	855	856
Old cetane index (JIS K 2204)		-	15	13.0	13.5	33.0	36.0	30.0	34.5	36.5
New cetane index (ISO 4264)		-	19.8	22.5	23.1	32.5	34.5	35.5	35.2	35.3
10%carbc	on residue	wt%	0.1	0.11	N/A	N/A	0.05	0.02	0.46	0.05
Sul	fur	wt%	0.14	0.13	0.39	0.049	0.058	0.047	0.14	0.073
Nitrogen		wt%	0.011	0.012	N/A	N/A	0.016	0.008	0.04	0.024
Carbon		wt%	90.2	89.9	N/A	N/A	88.6	88.8	88.4	88.8
Hydrogen		wt%	9.6	9.9	N/A	N/A	11.3	11.1	11.4	11.1
Molecular	Saturated molecule	vol%	14.3	16.7	14.6	35.0	39.8	39.2	45.4	44.8
structures	Olefin	vol%	1.5	16.8	0.8	7.0	6.2	4.5	3.0	7.4
	Aromatics	vol%	84.2	66.5	84.6	58.0	54.0	56.3	51.6	47.8
	1ring aromatics	vol%	36.6	33.2	16.5	21.8	20.7	34.1	17.9	12.8
	2ring aromatics	vol%	43.6	27.8	47.4	24.6	22.8	17.9	23.9	18.5
	over 2 ring aromatics	vol%	4.0	5.5	20.7	11.6	10.5	4.3	9.8	16.5

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166 2.3 Experimental setup and test conditions

Figure 2 shows the experimental setup 20 and the droplet suspend holder used in this study. The pressure vessel (inner diameter: 100 mm; inner height; 229 mm) was made by extra-super duralumin (JIS: A7075). The pressure vessel was filled with N₂ before the experiment to observe

170 a pure evaporation phenomenon without combustion. A charge-coupled device (CCD) video 171 camera and a high-speed camera (DITECT, HAS-X) was used to observe the inside of the pressure 172 vessel. The CCD video camera observed the droplet generating process. A fuel droplet with a 173 diameter in the range of $0.5 \pm 15\%$ mm was suspended on the intersection point of two Al₂O₃/SiO₂ 174 fibers with 7 µm diameter. The fuel was pumped with a microflow pump to the tip of the glass 175 needle (outer diameter: 40 µm) of the droplet generator. After the fuel droplet was suspended at 176 the intersection point of the droplet suspender, the glass needle was evacuated in preparation for 177 the droplet insertion of the droplet into the hot chamber. A linear slider-crank mechanism was 178 applied to the fuel droplet elevator to reduce the impact on the suspended droplet during the droplet 179 insertion process. The traveling time of the droplet during the insertion into the hot chamber was 180 0.26 s, whose traveling distance is 6 mm. We compared the measured droplet diameter before and



(a) Whole of experimental setup (b) Schematic of the droplet holder



181 after droplet insertion for all measured 182 experimental data in the case of L1 to clarify 183 the effect of the linear slider-crank mechanism 184 on the evaporation characteristics. As a result, 185 the average error of diameter difference is less 186 than 5%. The ambient temperature (T_{amb}) 187 inside the hot chamber was arbitrarily 188 controlled in the range of 473-873 K by a 189 thermal control unit. The images of an 190 evaporating droplet were taken with a high-



Figure 3. Schematic of the image analysis used to measure the droplet diameter

191 speed camera with backlit photography. The recorded backlit image of an evaporating fuel droplet was analyzed by using a self-made image analyzer program ³². Figure 3 shows the details of the 192 193 image analysis in this study. Each backlit image of a fuel droplet was divided into horizontal line 194 sections with a vertical thickness of 2 pixels. The average horizontal intensity profiles were 195 calculated for each section. The edge of a droplet was determined as the farthest position from the 196 suspender at which the intensity was less than $(I_A + 2I_B)/3$, where I_A and I_B were the average 197 intensities of the background and droplet, respectively. The droplet volume was calculated by an 198 ellipsoidal approximation using the determined edge. Finally, the droplet diameter was defined as 199 the diameter of a sphere with the same volume. The measurement limit of the droplet diameter 200 using the image analyzer was under 0.078 mm, which corresponded to D^2/D_0^2 of 0.025.

Figure 4 describes the definition of the periods during the droplet evaporation. The squared diameter was normalized by the squared initial diameter. Note that the droplet diameter when the fuel droplet was inserted in the hot chamber was defined as the initial droplet diameter in this study. 204 In the droplet evaporation experiments, the 205 droplet diameter initially increases because of 206 the thermal expansion effect caused by 207 momentarily increasing the inner droplet 208 temperature, which is shown as an initial 209 heating period (τ_i/D_0^2) . In the initial heating 210 period, most of the heat energy from the 211 ambient gas into the droplet is consumed to 212 heat up the droplet. Therefore, decreasing the



Figure 4. Definition of periods during the droplet evaporation

213 droplet density leads to an increase in the droplet diameter until the droplet evaporation rate 214 becomes equal to the thermal expansion rate. The droplet diameter decreases monotonically after 215 the droplet diameter reaches the maximum. As the indicator of the droplet lifetime, the 95vol% lifetime (τ_{95}/D_0^2) was defined as the period between the time when the fuel droplet was inserted 216 217 into the hot chamber and the time when the droplet volume decreased to 5% of the initial volume 218 equivalent to a relative diameter square of 13.6%. The droplet lifetime was also normalized by the 219 second power of the initial droplet diameter because the initial droplet diameter strongly affects 220 the droplet lifetime. The period obtained by removing the initial heating period from the 95vol% lifetime is defined as the main evaporation period (τ_{evap}/D_0^2) . 221

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239 240 241 order of L2, L12, and L3. The reason for the pressure dependence of L13 becoming 242 243 different from the other LCO at the lower 244 ambient temperature will be discussed in 245 Section 3.2. Figure 6 shows the τ_{95}/D_0^2 of the 246 arranged-fuels in the ambient temperature 247 range of 473-873 K at the ambient pressures of 0.1, 0.5, and 1.0 MPa. Similar to LCO, the 248 trend that the differences of τ_{95}/D_0^2 become



Figure 5. Normalized 95vol% lifetime (τ_{95} / D_0^2) as a function of the ambient temperature for LCO

250 large with decreasing ambient temperature can 251 be seen at all ambient pressures. On the other 252 hands, the differences of τ_{95}/D_0^2 between S30 253 and S40 become much small at various 254 ambient temperatures, except 473 K, at all 255 ambient pressures.

The reason that the differences of τ_{95}/D_0^2 256 between the fuels become large at the low 257 258 ambient temperature will be discussed in the 259 parts that follow. Figure 7 shows the histories of the D^2/D_0^2 of LCO and the arranged-fuels 260 261 at various ambient temperatures and $P_{amb} =$ 262 0.1 MPa. Note that the detailed law data for all histories of the D^2/D_0^2 obtained in this study 263 are provided in the Supporting Information 264 section of this paper. These D^2/D_0^2 history 265 data will be directly used for the future 266 267 validation for the multi-component droplet 268 evaporation model development. In particular, 269 the sensitivity of the components in fuel for the 270 models can be validated in detail, because the D^2/D_0^2 histories significantly differ each other 271 272 when the components are different as



Figure 6. Normalized 95vol% lifetime (τ_{95}/D_0^2) as a function of the ambient temperature for the arranged-fuel

273 observed in Figure 7 (a). According to Figure 7-(a), the rate of decrease in D^2/D_0^2 274 275 become obviously slow as time goes by. The rate of decrease in D^2/D_0^2 approaches to a 276 during τ_{evap}/D_0^2 with 277 constant rate 278 increasing ambient temperature (Figure 7). 279 To evaluate the rate of decrease in D^2/D_0^2 , 280 this study defined the instantaneous 281 evaporation coefficient (K_{inst}) to 282 quantitively evaluate the rate of decrease in D^2/D_0^2 . K_{inst} was calculated by the 283 284 following procedures: first, the original data plots during τ_{evap}/D_0^2 consisting of 200-285 286 3000 plots were thinned to 40-50 plots. 287 Second, the smoothing differentiation method, called as the SG method ³³, was 288 289 applied to the thinned data plots. The 290 absolute value of the differential coefficient 291 calculated by the SG method gave K_{inst} . Figure 8 shows the K_{inst} of LCO and the 292 293 arranged-fuels ambient in various temperatures at $P_{amb} = 0.1$ MPa. The 294 fluctuation of K_{inst} history shown in Figure 295



Figure 7. Examples of the histories of normalized D^2 for LCO

296 8 is caused by image analysis error. A decreasing of K_{inst} indicates that the rate of 297 decrease in D^2/D_0^2 shown in Figure 7 is not 298 299 constant as time goes by at T_{amb}=473 K. The 300 gradient shown in Figure 8-(a) become steep 301 in the order of S30, L12, L13, and L1. This 302 order matches the order of the highest boiling 303 point component in the fuel. Hence, it is considered that the end point of distillation 304 305 curve may make the rate of decrease in D^2/D_0^2 slower even though LCO contain the 306 307 various chemical species having the various 308 boiling point. Note that there is an obvious difference of Kinst between L12 and L13 309 310 even though the difference of the highest 311 boiling point component between L12 and 312 L13 is much small. To clarify this reason, 313 much fundamental experimental data based 314 on the single component are necessary. 315 However, Figure 8-(b) and (c) shows that the difference of Kinst between the fuels 316 decrease, and the absolute value of K_{inst} 317 318 reaches an almost constant value with



Figure 8. Instantaneous evaporation coefficient (K_{inst}) for LCO and the arranged-fuels

increasing ambient temperature. These findings confirm that the rate of decrease in D^2/D_0^2 is almost constant at the higher ambient temperature. On the other hand, the ambient pressure dependence on the evaporation characteristics will be discussed in section 3.3.

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Molecular Formula $C_{10}H_{14}$ C11H10 C16H34 C20H42 Molecular Weight g/mol 134 142 226 282 Mass Fraction 0.3116 0.4402 0.1753 0.0729 °C **Boiling Point** 169.1 244.6 286.9 344.1 Density (15°C) g/cm³ 0.8708 1.027 0.7773 0.7908

Table 2. Compositions and properties of L1 surrogate fuel

323 3.2 Development and validation of the surrogate fuel for LCO

In the numerical simulation, a surrogate fuel is usually used. A surrogate fuel consists of distinct fuel to simulate a certain characteristic of physical phenomena. By using a surrogate fuel in combustion simulation, we are able to utilize the detailed chemical reaction mechanism. However, the surrogate fuel of LCO has not been developed. To conduct the precise combustion simulation using LCO in future works, the surrogate fuel of LCO should be developed.

To simulate the evaporation characteristics of LCO (L1), we choose the chemical species and determine their fractions so that the distillation characteristics of LCO can be reproduced by the surrogate fuel. Table 2 shows the compositions and properties of the developed surrogate fuel. LCO surrogate fuel, which is treated as a mixture composed of tert-butylbenzene ($C_{10}H_{14}$), 1methylnaphthalene ($C_{11}H_{10}$), n-hexadecane ($C_{16}H_{34}$) and n-eicosane ($C_{20}H_{42}$) with mass fraction of 0.3116, 0.4402, 0.1753, and 0.0729, respectively, was developed. The boiling point with volume

Fuel		L1	Surrogate Fuel
Density (15°C)	g/cm ³	0.9323	0.9173
Cetane Index [JIS K 2280]		19.8	19.7
Sulfur	wt%	0.14	0
Nitrogen	wt%	0.011	0
Carbon	wt%	90.3	89.9
Hydrogen	wt%	9.6	10.1
C/H ratio	-	9.4	8.9
Saturated molecule	vol%	14.3	16
Olefin	vol%	1.5	0
Aromatics	vol%	84.2	84
1ring aromatics	vol%	36.6	36
2ring aromatics	vol%	43.6	48
over 2 ring aromatics	vol%	4	0

Table 3. Properties of L1 surrogate fuel

fraction for each chemical species was compared with the distillation curve of L1. The boiling temperature width of the surrogate fuel stays within the boiling temperature range of LCO, especially the first drop point and ending point of L1 distillation curve is in good agreement with

338 the boiling point of components in the 339 surrogate fuel. Second, the properties of the 340 surrogate fuel are compared with that of LCO. 341 According to table 3, the surrogate fuel emulates the density at 15 $^\circ$ C , the NCI 342 343 calculated by eq 4, the C/H ratio, and the 344 molecular types of L1. Hence, the 345 thermodynamic properties and the ratio of 346 molecular structure types of the surrogate fuel



Figure 9. Validation of LCO surrogate fuel at various ambient temperature.

347 are in good agreement with those of L1. Finally, the droplet evaporation experiments of the surrogate fuel were conducted in the present set-up to validate the evaporation characteristics. 348 Figure 9 shows the comparison of droplet lifetimes between the surrogate fuel and L1 at various 349 350 ambient temperatures. It is found that the droplet lifetimes of the surrogate fuel are in good 351 agreement with that of L1 at various ambient temperatures. Especially, the difference of droplet 352 lifetime between L1 and the surrogate fuel was less than 5% in higher ambient temperature. It can 353 be concluded that we succeeded to develop the LCO surrogate fuel, which well simulates the 354 droplet evaporation characteristics of LCO.

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356 3.3 Effect of the ambient pressure on the evaporation characteristics for LCO and arranged-fuels.

357 Figure 10 shows the pressure dependence of τ_{95}/D_0^2 for L1, L2, and L13 at $T_{amb} =$ 358 473 and 873 K. When $T_{amb} = 873 K$, the 359 difference of τ_{95}/D_0^2 between the fuels 360 become smaller at all ambient pressure. On the 361 362 other hands, when $T_{amb} = 473 \text{ K}$, the difference of τ_{95}/D_0^2 between the fuels 363 364 become large at all ambient pressures. Accordingly, the τ_{95}/D_0^2 of L13 is larger than 365 366 that of L1 and smaller than that of L2 at



Figure 10. Normalized 95vol% lifetime (τ_{95}/D_0^2) as a function of the ambient pressure for LCO

367 $T_{amb} = 473$ K and $P_{amb} = 0.1$ MPa. However, the τ_{95}/D_0^2 of L13 steeply increase with increasing 368 ambient pressure (Figure 10). Eventually, the τ_{95}/D_0^2 of L13 becomes the largest among LCO at 369 P_{amb} = 1.0 MPa. Figure 10 depicts the τ_{95}/D_0^2 370 become large in the order of increasing the 371 ending point of the distillation curve shown in 372 Figure 1.

373 Figure 11 shows the relationships between 374 the distillation temperature and τ_{95}/D_0^2 at 375 various ambient temperature at $P_{amb} = 0.1$ and 376 1.0 MPa. In both figures (Figure 11), the left 377 region shows the range of 10vol% distillation 378 temperature for all species, whereas the right 379 region illustrates the range of 90vol% 380 distillation temperature for all species. The scattering of τ_{95}/D_0^2 is smaller at the high 381 382 ambient temperature at various ambient 383 pressures. In other words, the droplet lifetimes 384 independent of the distillation are 385 temperatures at the high ambient temperature 386 at various ambient pressures. However, τ_{95} /



Figure 11. Normalized 95vol% lifetime (τ_{95}/D_0^2) as a function of the distillation temperature for the fuels used in this study

387 D_0^2 become longer with increasing 90vol% distillation temperatures in low ambient temperatures. 388 In Figure 11, linear least-squares fitting equations for T_{amb} = 473 K are shown. Apparently, the 389 gradient of the fitting line becomes steeper in higher ambient pressure. This means that the 390 relationships between 90vol% distillation temperature and droplet lifetime become strong in low ambient temperature and high ambient pressure. The reason for this relationship will be discussedin the paragraphs that follow.

We conducted a Peclet number analysis to discuss the above relationship. According to ref. ³⁴, the molar weight variation has a decisive influence on the ratio of the characteristics time scales of the droplet lifetime and the mass diffusion. The gasification Peclet number ³⁵ shown in eq 6 can assess the relative dominance of the diffusion limit versus distillation behavior.

397

$$Pe = \frac{\dot{m}_{vap}}{2\pi D_d \Gamma_l \rho_l} \tag{6}$$

398

where *Pe* is the Peclet number (-), \dot{m}_{vap} is the mass evaporation rate (kg/s), D_d is the droplet diameter (m), Γ_l is the diffusion coefficient inside the droplet (m²/s), and ρ_l is the droplet density (kg/m³). However, the Peclet number given by eq 6 is not considering the multi-component interaction effect. Therefore, the Peclet number on droplet evaporation was estimated in this paper by rearranging eq 6 to multi-component fuel.

404 The Peclet number shown in eq 6 can be rearranged by using the Spalding model ³⁴. Mass 405 evaporation rate (\dot{m}_{vap}) and evaporation constant (*K*) is described as the following.

406

$$\dot{m}_{vap} = \frac{4\pi k_g r_s}{c_{p,g}} \ln \left(B_q + 1\right) \tag{7}$$

$$K = \frac{8k_g}{\rho_l c_{p,g}} \ln \left(B_q + 1 \right) \tag{8}$$

407

Where k_g is the thermal conductivity of ambient gas (W/m-K), $c_{p,g}$ is the specific heat of gas (J/kg-K), r_s is droplet radius (m), ρ_l is fuel density (kg/m³), B_q is Spalding number (-). Substituting eq 8 into eq 7, the mass evaporation rate can be expressed as a function of evaporation constant (*K*), and the Peclet number shown in eq 6 can be rearranged as the following.

412

$$Pe = \frac{\dot{m}_{vap}}{2\pi D_d \Gamma_l \rho_l} = \frac{1}{4\pi r_s \Gamma_l \rho_l} \frac{\pi r_s \rho_l}{2} K = \frac{K}{8\Gamma_l}$$
(9)

413

414 Note that the surrogate fuel developed in section 3.2 was used to calculate the diffusion coefficient 415 inside the droplet (Γ_l). In, our analysis, *K* was replaced by instantaneous evaporation coefficient, 416 K_{inst} , which was obtained from experimental results shown in Figure 8.

Figure 12 shows the history of estimated Peclet number at two ambient conditions, (1) $T_{amb} =$ 417 473 K and $P_{amb} = 1.0$ MPa (2) $T_{amb} = 873$ K and $P_{amb} = 0.1$ MPa. Pe shown in Figure 12 are 418 419 calculated by eq 9. Furthermore, K_{inst} used in eq 9 was obtained from experimental results. This 420 is why Pe shown in Figure 12 is renewed moment by moment. Note that the diffusion coefficient 421 inside the droplet (Γ_l) is a function of droplet temperature (Appendix). As a droplet temperature, 422 473 K which is the same temperature with ambient temperature was applied in case (1), and 617 423 K which is the same temperature with the boiling point of n-eicosane was applied in case (2). 424 According to Figure 12, the Peclet number in case (1) decreases as time goes by although the Peclet 425 number in case (2) shows almost constant value, and the Peclet number in case (1) becomes much 426 smaller than that in case (2). When the Peclet number becomes larger, the internal diffusion is not 427 able to catch up with the mass evaporation rate. Therefore, the droplet composition remains unchanged ³⁴, thus indicating that evaporation characteristics approach the evaporation 428 characteristics of the single component described by the D² law. This is why the correlation 429

430 between the droplet evaporation 431 characteristics and the distillation process is 432 weak at high ambient temperatures. On the 433 other hand, when the Peclet number becomes 434 smaller, the mass evaporation rate becomes 435 sufficiently slow compared with the internal 436 diffusion. This result implies that the 437 relationship between the droplet evaporation 438 characteristics and the distillation curve is strong. This is why the D² law cannot be 439 applied to the evaporation of multi-component 440 441 fuel droplet at the low ambient temperature. 442 Moreover, the evaporation rate is further reduced at high pressure ambient. Therefore, 443 the tendency of droplet lifetime lengthening 444 with increasing the ending point of the fuel 445 446 distillation temperature become strong under 447 the low ambient temperature and high ambient 448 pressure.



Figure 12. Histories of estimated Peclet number during main evaporation term



Figure 13. Normalized 95vol% lifetime (τ_{95}/D_0^2) as a function of the ambient pressure for the arranged-fuels

449 The relationships between the different ways of calculating the cetane index and droplet 450 evaporation characteristics were investigated. Figure 13 shows the pressure dependence of τ_{95}/D_0^2 451 for the arranged-fuels. As mentioned in 452 Section 2.2, the arranged-fuels were made 453 such that their NCI is 35. Figure 13 shows that the difference of τ_{95}/D_0^2 between the fuels 454 became small at $T_{amb} = 673,773$, and 455 873 K. However, the difference of τ_{95}/D_0^2 456 457 between the fuels became large at $T_{amb} =$ 473 and 573 K. As discussed in Section 3.1, 458 459 this behavior is caused by components with a 460 high boiling point, which make the rate of decrease in D^2/D_0^2 slower. By contrast, the 461 OCI of S30, S35, and S40 are increased in the 462 463 order of S30, S35, and S40. However, the τ_{95}/D_0^2 of S30, S35, and S40 is increased in 464 the order of S30, S40, and S35. These results 465 implied that the τ_{95}/D_0^2 is independent of the 466 OCI. Simultaneously, the τ_{95}/D_0^2 of S30, S35, 467 468 and S40 increased in the order of S30, S40, 469 and S35, although all the arranged-fuels have 470 the almost same new cetane index that is 35. In conclusion, τ_{95}/D_0^2 cannot be associated 471 472 with the cetane index when the cetane index is 473 35.



Figure 14. Normalized 95vol% lifetime (τ_{95}/D_0^2) as a function of the cetane index for the fuels used in this study

474 Finally, the relationships between the droplet lifetime and the cetane index were investigated. Figure 14 shows the relationships between the cetane index calculated by eq 4 and τ_{95}/D_0^2 at the 475 476 various ambient pressures (0.1, 0.5, and 1.0 MPa) and temperatures. It was found that there is no correlation between the cetane index and τ_{95}/D_0^2 . It can be concluded that the droplet lifetime is 477 478 independent of the cetane index introduced in this paper under the conditions tested. Furthermore, 479 the correction coefficient between the droplet evaporation lifetime and the mole fraction of mono-, 480 di-, and 3+ cyclic aromatic compounds at all experimental conditions were investigated. However, 481 we found that there is little correlation between the evaporation characteristics and aromatic 482 compositions. In actuality, fuel droplets will continuously evaporate at higher temperature and 483 pressure conditions after the ignition. Therefore, the clarification of droplet evaporation 484 characteristics in such conditions is also necessary as the future research.

485

486 4. CONCLUSIONS

487 In this study, LCO single droplet evaporation experiments were conducted at various ambient 488 temperature and pressure to obtain the validation data. The detailed droplet evaporation history 489 data of fuels with various composition ratios were obtained by this study. The data can be utilized 490 for the validation of the sensitivity of the components for multi-component droplet evaporation 491 models in the future. In addition, development and validation of the surrogate fuel for LCO (L1) 492 were conducted. Finally, the relationships between the evaporation characteristics and cetane index, 493 which is one of the indicators for fuel ignitability, were investigated. The main findings are as 494 follows:

The droplet lifetime decreases with increasing ambient temperature for all fuel species. The
 differences of droplet lifetime between the fuels become larger with decreasing ambient

- 497 temperature. This is because the low volatile component made the evaporation rate498 outstandingly slow at low ambient temperature.
- 2. The evaporation characteristics of the surrogate fuel are in good agreement with that of LCO
 at various ambient temperatures. Especially, the difference of droplet lifetime between L1 and
 the surrogate fuel is less than 5% in higher ambient temperature.
- 502 3. The droplet evaporation lifetime can be characterized by the late stage distillation temperature 503 at a low ambient temperature. The correlation between the droplet lifetime and the late-stage 504 distillation temperature becomes stronger with increasing ambient pressure because the mass 505 evaporation rate becomes smaller than the internal diffusion, which is a condition similar to 506 that in the distillation test. Hence, the tendency of the droplet lifetime to increase with 507 increasing the end point of the fuel distillation temperature become strong at low ambient 508 temperature and high ambient pressure.
- 509 4. The droplet lifetime of LCO and the arranged-fuels is independent of the cetane index under510 experimental conditions shown in this paper.
- 511
- 512 Appendix Diffusion coefficient inside a droplet
- 513 The diffusion coefficients used in the Peclet number were estimated as the following. First, the 514 binary diffusion coefficient can be calculated from ref. ³⁶ as
- 515

516
$$D_{i,j} = 8.93 \times 10^{-8} \frac{V_j^{0.267}}{V_i^{0.433}} \frac{T}{\eta_j}$$

517

where $D_{i,j}$ is the binary diffusion coefficient of species *i* and *j* in the droplet; *V* is the molar volume of species *i*; *T* is droplet temperature; η_j Pearson's the viscosity of species *i*. For multicomponent fuels, the mixture of all the species except *i* is assumed as the second species *m*. Then, the diffusion coefficient of species *i* for the mixture can be described as

523
$$D_{i,m}\eta_m^{0.8} = \sum_{\substack{j=1\\ i\neq i}}^{N_l} x_j D_{i,j}\eta_j^{0.8}$$

524

525 where x_j is the mole fraction of species *j* and N_l is the number of species. Hence, the binary 526 diffusion coefficient (Γ_l) used for Peclet number in eq 9 can be averaged as the following.

527
$$\Gamma_l = \sum_{i=1}^{N_l} x_i D_{i,m}$$

528

529

- 530 **ASSOCIATED CONTENT**
- 531 Supporting Information.
- 532 The following files are available free of charge.
- 533 Experimental data_LCO (xlsx)

534

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538 Notes

- 539 The authors declare no competing financial interest.
- 540
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