

1 **Optical diagnostic study of internal and external EGR combined with oxygenated**
2 **fuels of n-butanol, POE₃ and DMC to optimize the combustion process of FT**
3 **synthetic diesel**

4

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10

11 **Abstract**

12 With the introduction of carbon neutrality target, Fischer-Tropsch (FT) synthetic
13 fuels are coming back into the limelight as a kind of carbon-neutral fuel. However, the
14 mismatch between the overly high cetane number (CN) and the relatively low
15 vaporability of FT synthetic diesel is unfavorable to the soot emission control, which
16 will make it difficult to meet more stringent fuel consumption and emission regulations
17 in future applications. To investigate the potential of oxygenated fuels combined with
18 different exhaust gas recirculation (EGR) introduction schemes to achieve high-
19 efficiency and clean combustion of FT synthetic diesel, an optical diagnostic study was

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20 carried out based on high-speed photography and the two-color method. The results
21 show that all three kinds of oxygenated fuels could suppress soot emissions via self-
22 carrying oxygen and adjusting the physicochemical properties of the fuel blend.
23 Compared with the combustion characteristics of FT synthetic diesel, the flame area
24 and luminosity of oxygenated blends are reduced, and the in-cylinder temperature and
25 soot KL factor are lowered. Among them, n-butanol exhibits a greater capability of soot
26 control compared to polyoxymethylene dimethyl ethers (PODE₃) and dimethyl
27 carbonate (DMC). In addition, introducing internal and external EGR to the engine
28 fueled by n-butanol/FT synthetic diesel blend shows that with the increase of EGR rate,
29 the external EGR exhibits a gradually stronger inhibiting effect on the heat release
30 process and soot KL factor, while the internal EGR exhibits an inhibiting and then
31 promoting effect. Moreover, the high ratio internal EGR shortens the ignition delay (ID)
32 significantly due to the strong heating effect, which is unfavorable to the control of soot
33 emission. The combination of oxygenated fuels and internal/external EGR could
34 effectively optimize the combustion process of FT synthetic diesel and inhibit soot
35 generation, but the EGR rate needs to be controlled within a proper range.

36 *Keywords:* FT synthetic diesel; N-butanol, PODE₃, DMC; Flame characteristics; Two-
37 color method; Internal and external EGR

38 **Highlights:**

- 39 ● An optical diagnostic of optimizing FT synthetic diesel combustion was conducted.
40 ● Blending oxygenated fuels contribute to combustion optimization and soot
41 reduction.

- 42 ● N-butanol exhibits the strongest soot-inhibiting ability followed by PODE₃ and
 43 DMC.
- 44 ● External EGR exhibits an inhibiting effect on combustion and diffusion flame.
- 45 ● Heating effect of high-rate internal EGR neutralizes its inhibiting effect.

Abbreviations			
FT	Fischer-Tropsch	LII	Laser-induced incandescence
CN	Cetane number	PIV	Particle image velocimetry
CI	Compression ignition	PDPA	Phase doppler particle analyzer
ITE	Indicated thermal efficiency	ECU	Electronic control unit
COV	Coefficient of variation	MCU	Microprogrammed control unit
PPRR	Peak pressure rise rate	VVT	Variable valve timing
PHRR	Peak heat release rate	HRR	Heat release rate
R _p	Premixed gasoline ratio	IMEP	Indicated mean effective pressure
ID	Ignition delay	CR	Compression ratio
DMC	Dimethyl carbonate	RGB	Red green blue
PODE	Polyoxymethylene dimethyl ethers	SINL	Spatially integrated natural luminosity
EGR	Exhaust gas recirculation	ϕ_a	Excess air ratio
NVO	Negative valve overlap	SOI	Start of injection
ICE	Internal combustion engine	NVOA	Negative valve overlap angle
LIF	Laser-induced fluorescence	CD	Combustion duration

46 **1 Introduction**

47 Global warming is a worldwide problem for mankind. Throughout more than 150
 48 years, human activities have produced a considerable amount of greenhouse gases into
 49 the atmosphere, causing significant impacts on the climate ^[1-3]. To ensure the
 50 sustainability of ecosystems and human society, the target of limiting global warming
 51 to 1.5°C has been proposed ^[4,5], which calls for halving carbon emissions by 2030 and

52 achieving carbon neutrality by 2060 ^[6-8]. In this regard, the development of clean
53 low/zero carbon fuels plays an important role in the decarbonization of the energy and
54 transportation sectors ^[9,10], such as biofuels based on direct photosynthesis ^[11,12], green
55 hydrogen/green ammonia ^[13,14], and e-fuels based on green electricity, etc ^[15,16].
56 Although Fischer-Tropsch (FT) synthetic diesel still has problems such as high pre-
57 investment costs compared with other alternative fuels ^[17], it also has some unique
58 advantages such as high energy density, greater compatibility with existing storage and
59 refueling infrastructure, and adaptability to current engine technology ^[18].

60 Compared with conventional petroleum diesel, FT synthetic diesel has a higher
61 content of straight-chain alkanes, lower sulfur, and aromatic hydrocarbons, higher
62 cetane number (CN), better flammability, and can be miscible with conventional diesel
63 in any ratio. These features make FT synthetic diesel a promising alternative fuel for
64 compression ignition (CI) engines ^[19-21]. Many studies have shown that the use of FT
65 synthetic diesel contributes to the optimization of combustion and emission
66 characteristics of CI engines. Shi et al. studied the effect of FT synthetic diesel fuel on
67 a turbocharged, intercooling, common-rail diesel engine and showed that FT synthetic
68 diesel fuel contributed to a higher indicated thermal efficiency (ITE) and lower
69 coefficient of variation (COV) compared to China VI diesel ^[22]. Zhang et al. compared
70 the impacts of using diesel and FT synthetic diesel as pilot fuel on combustion and
71 emission characteristics respectively in RCCI mode ^[23]. It showed that FT synthetic
72 diesel with higher CN could reduce COV, control the peak pressure rise rate (PPRR)
73 and peak heat release rate (PHRR), and expand the load range of RCCI mode under a

74 low premixed gasoline ratio (R_p). In addition, the authors pointed out that the FT
75 synthetic diesel fuel could reduce soot emissions, and similar conclusions were
76 mentioned in several other studies [24-28].

77 The lower soot emissions of FT synthetic diesel are mainly due to the low aromatic
78 content, which inhibits the formation of soot precursors [25,26]. However, with the
79 improvement of commercial fuel standards, the aromatic content of fossil diesel will be
80 gradually reduced, and the advantage of low aromatic content of FT synthetic diesel
81 will gradually disappear, while its higher CN will lead to worse combustion timeliness
82 and higher soot emission [29,30]. Adding oxygenated fuels with different molecular
83 structures such as n-butanol [31,32], dimethyl carbonate (DMC) [33,34], and
84 polyoxymethylene dimethyl ethers (PODE) [35,36] could exert the advantage of the
85 oxygen-carrying property on inhibiting the generation of soot [37-39]. Oxygenated
86 additives are capable of all-round combustion regulation from the physicochemical
87 properties of the fuel, such as ignition and combustion phases, mixture stoichiometry
88 distribution, adiabatic flame temperature, and temperature field distribution of the
89 combustion process, etc. [40-42], so it is necessary to investigate the effect of the
90 introduction of oxygenated fuels on the combustion process of FT synthetic fuel.
91 Additionally, as a means of adjusting the properties of working medium, EGR also
92 plays an important role in the combustion regulation [43-45]. Apart from external EGR
93 which is used commonly, internal EGR is also an effective method of combustion
94 control. Duan et al. implemented internal EGR through negative valve overlap (NVO)
95 and investigated the effect of internal EGR on combustion and emissions of the engine

96 [46]. The results showed that NO_x emissions were reduced but the CO and HC emissions
97 increased as the internal EGR rate increased. It is necessary to carry out research on
98 controlling the combustion process of FT synthetic diesel fuel by combining
99 internal/external EGR with oxygenated fuels, however, relevant studies have been
100 rarely reported.

101 As a non-intrusive research method, optical diagnostic has the characteristics of
102 fast response, high sensitivity, and intuitive results, thus becoming an important method
103 in the study of the working process of the internal combustion engine (ICE), such as
104 fuel injection, vaporation, air-fuel mixing, flame development, and pollutant generation
105 history, etc [47-49]. Recently, with the advancement of optics and materials science, a
106 variety of optical diagnostic methods such as laser-induced fluorescence (LIF), laser-
107 induced incandescence (LII), particle image velocimetry (PIV), Phase Doppler Particle
108 Analyzer (PDPA) and high-speed photography have also been used in the research of
109 ICE widely [29,50-55]. Matsui et al. introduced an optical diagnostic technique of high-
110 speed photography combined with the two-color method based on solid thermal
111 radiation theory [56,57], which has been improved by the researchers and is now capable
112 of systematically studying the flame development history, in-cylinder temperature field,
113 and soot concentration field [58-62]. Jeon et al. investigated the flame characteristics,
114 temperature field, and soot KL factor field during the combustion of a biodiesel/diesel
115 blend (biodiesel at 20% v/v) at various injection pressures based on the two-color
116 method [63]. The results showed that the flame temperature and soot concentration of
117 the oxygenated blend were higher at all the injection pressures compared to those of

118 pure diesel. Therefore, optical diagnosis is an essential and effective research method
119 in revealing the impacts of oxygenated fuels and internal/external EGR on the
120 combustion and the soot generation processes during the combustion of FT synthetic
121 diesel.

122 In this study, an optical diagnostic test system was built based on a single-cylinder
123 diesel engine with flexible and adjustable combustion boundary conditions. The effects
124 of oxygenated fuel combined with internal/external EGR on the combustion process
125 and soot generation history were investigated using FT synthetic diesel as the base fuel.
126 The results will be able to establish a theoretical basis for the development of high-
127 efficiency and clean combustion strategies based on FT synthetic diesel.

128 **2 Experimental setup and test procedure**

129 *2.1 Optical Engine Test Bench*

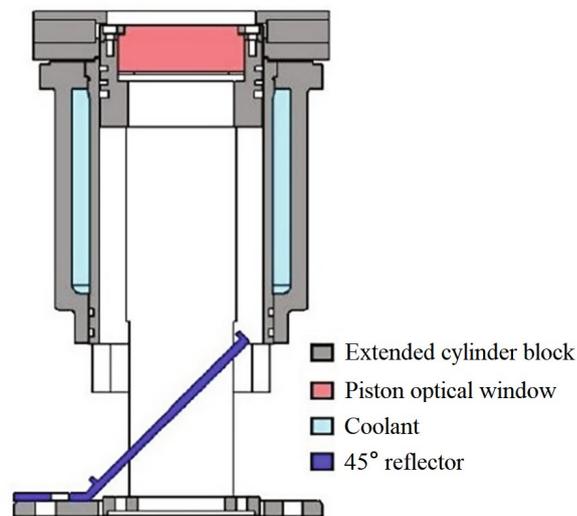
130 **Fig. 1** shows the layout of the optical engine measurement and control platform.
131 The test platform consisted of optical engine, electronic control system and high-speed
132 photography system. The optical engine was driven by an electric motor to operate
133 steadily at a constant speed, and the engine position signal was acquired by the
134 crankshaft and camshaft position sensors and transmitted to the control system (open
135 electronic control unit (ECU) NI 2106). The NI system took the control of injection
136 parameters and sent the trigger signal to the microprogrammed control unit (MCU)
137 once per cycle. The MCU implemented the control of the high-speed camera and the
138 hydraulic variable valve timing (VVT) system. Moreover, the real-time acquisition and
139 analysis of the combustion state were carried out by the combustion analyzer based on

156 the lowered 45° reflector. The structure of the extended cylinder and the schematic of
 157 the optical pathway are shown in **Fig. 2**. In addition, to implement internal EGR through
 158 flexible control of valve timing, a hydraulic VVT system was adopted to replace the
 159 stock valvetrain. The original compression ratio (CR) of the optical engine is 17. Due
 160 to the shape requirements of optical glass, the piston head could only take the shape of
 161 a flat top. The flat piston head limited the CR of the optical engine to a maximum of 13.
 162 To ensure a reliable compression ignition, the temperatures of the intake air and the
 163 coolant were heated to 353 K and 363 K respectively.

164 **Table 1** Technical specifications of the optical engine.

Category	Properties
Bore×stroke / mm	105×114.3
Connecting rod length / mm	190
Displacement / L	0.99
Geometric compression ratio	13
Valve lift / mm	11
Number of valves	4
Number of injector nozzle holes	7
Diameter of nozzle hole / mm	0.12

165



166

167

Fig. 2. Schematic of optical pathway.

168 *2.2 High-speed photography and image processing*

169 The high-speed camera adopted in this study was Phantom v611 produced by
170 AMETEK, USA. The high-speed camera was operated under the external trigger mode,
171 based on the trigger signal from ECU and the synchronization signal from the encoder
172 to accomplish the accurate control of the sampling time. The sampling parameters of
173 the high-speed camera are shown in **Table 2**. The natural luminescence signal of the in-
174 cylinder flame captured under such sampling parameters mainly consisted of
175 narrowband chemiluminescence and broadband soot incandescence. The intensity of
176 chemiluminescence in the visible and near-infrared bands was much lower than that of
177 soot incandescence ^[65], so the in-cylinder flame images could be considered as soot
178 incandescence during diffusion combustion.

179 **Table 2** The sampling parameters of high-speed camera

Category	Properties
Resolution	512×512
Aperture level	7
Exposure time / μs	4
Sampling interval	1°CA

180 The raw images captured by the high-speed camera were saved in Red Green Blue
181 (RGB) format, and the flame images for analysis were pre-processed by cropping,
182 denoising, and enhancing the raw images via a self-programmed MATLAB script. Each
183 flame image was further processed by grayscale and binarization to identify the flame
184 region, then the percentage of flame area and the flame luminosity (spatially integrated
185 natural luminosity, SINL) were calculated ^[66,67]. In addition, the in-cylinder temperature
186 field and soot KL factor field were obtained by the two-color method. The two-color
187 method is a temperature measurement methodology developed on the basis of solid

188 thermal radiation theory ^[68,69]. The transcendental equation of the two-color method
 189 was solved by Newton's iterative method, and the corresponding MATLAB script was
 190 programmed to implement batch post-processing of flame images.

191 The temperature measurement via the two-color method necessitated a brightness
 192 temperature calibration with a high-temperature blackbody furnace to establish the
 193 correspondence between the RGB values of the image and blackbody temperature. The
 194 calibration images at different temperatures are shown in **Fig. 3**. The RGB values of
 195 each pixel within the image were extracted and averaged to obtain the calibration data
 196 at each temperature, as shown in **Table 3**. To validate the reliability of the calculation
 197 procedure, the calibration images were computed and compared with the actual
 198 temperature. The computational errors at each temperature are shown in **Table 4**,
 199 ensuring the accuracy of the calculation procedure employed in this study.

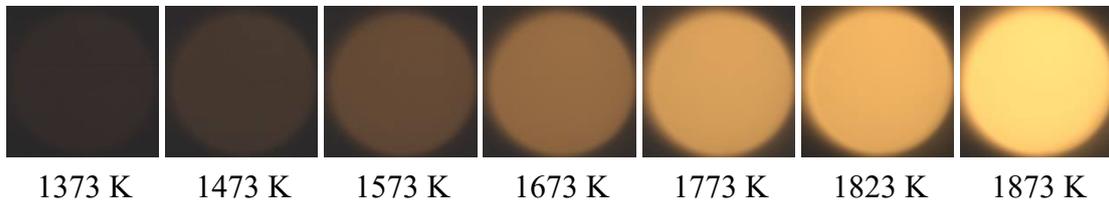


Fig. 3. The calibration images at various temperatures.

Table 3 The calibration data at various temperatures.

Temperature / K	R	G	B
1373	52.53	45.97	43.03
1473	70.28	55.95	47.07
1573	102.3	75.52	54.35
1673	150.71	109.11	67.81
1773	219.09	161.87	92.22
1823	240.3	179.54	98.48
1873	255	219.87	121.87

Table 4 The computational errors at various temperatures

Actual value / K	1373	1473	1573	1673	1773	1823	1873
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Estimated value / K	1388	1468	1574	1682	1787	1815	1864
Absolute error / K	15	-5	1	9	14	-8	-9
Relative error / %	1.09	-0.34	0.06	0.54	0.79	-0.44	-0.48

203 2.3 Experimental plan

204 In this experiment, FT synthetic diesel was used as the baseline fuel which was
205 labeled as FT. To investigate the effects of different oxygenated fuels on combustion
206 and soot generation process of FT synthetic diesel, several FT/oxygenated fuel blends
207 with the same oxygen content of 7.73% were adopted to ensure that the fuel-bound
208 oxygen content would not affect the test results ^[70]. The chosen oxygenated fuel
209 additives were DMC (C₃H₆O₃), PODE₃ (C₅H₁₂O₄) and n-butanol (C₄H₁₀O) with the
210 blending mass fractions of 14.5%, 19.3%, and 35.8%, the oxygenated blends were
211 labeled as DF, PF, and BF, respectively. In the following section, the potential of
212 oxygenated fuels coupled with internal/external EGR to achieve the clean combustion
213 of the CI engine was investigated based on BF. The main physicochemical properties
214 of the test fuels are listed in **Table 5**. The heat value of the fuel being injected into the
215 cylinder per cycle in this study was consistently controlled at 1438 J/cycle, with an
216 IMEP of approximately 0.4 MPa. During the experiment, the engine speed was
217 maintained at 1000 r/min, the intake mass flow and intake pressure were kept at 60 kg/h
218 ($\phi_a \approx 4$) and 1.8 bar (absolute pressure). The injection pressure was controlled at 100
219 MPa, and the start of injection (SOI) timing was fixed at -9°CA ATDC. The
220 correspondence between injection mass and injection pulse width for each test fuel was
221 calibrated before the experiment.

222 **Table 5** Physicochemical properties of test fuels

Category	FT	N-butanol	DMC	PODE ₃
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Density at 20°C / (kg·m ⁻³)	757	805.5	1069	1024.2
CN	75.4	25	35	78
Lower heating value / (MJ·kg ⁻¹)	43.07	33.1	13.5	19.05
Latent heat of vaporization / (kJ·kg ⁻¹)	~	585	369	~359 ^a
Kinematic viscosity at 25°C / (mm ² ·s ⁻¹)	2.14	3.64	0.63	1.05
Boiling point / °C	257.8	117.7	90	156
Total aromatics / %	0.8	0	0	0
Sulfur content / 10 ⁻⁶	0.38	0	0	0

223 ^a Taking the latent heat of vaporization of PODE₃₋₈ as the reference value ^[71]

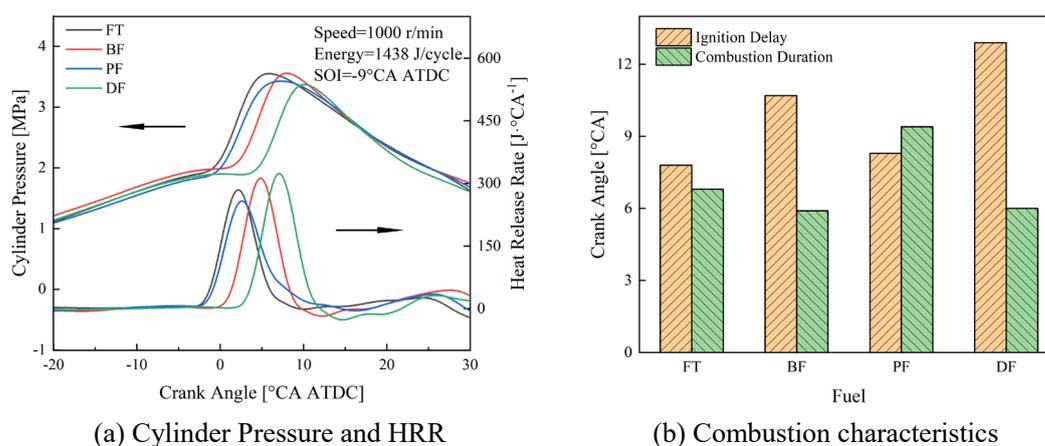
224 In this study, pure CO₂ was introduced through a gas cylinder as the simulated
 225 external EGR and the CO₂ gas was heated together with the intake air in the inlet. The
 226 selected external EGR rates in the experiment were 10%, 20%, and 30%, which were
 227 adjusted through the EGR regulating valve and air flow regulating valve. The NVO
 228 strategy was used to retain exhaust gases and thus implement internal EGR, and the
 229 valve timing was flexibly adjusted to achieve early exhaust valve closing and late intake
 230 valve opening. The selected negative valve overlap angles (NVOAs) in the experiment
 231 were 40°CA, 60°CA, 80°CA, 100°CA, and 120°CA, and the original NVOA of -28°CA
 232 was also taken into consideration.

233 **3 Results and discussions**

234 *3.1 Effects of oxygen-containing functional group structures on the FT synthetic diesel* 235 *combustion and soot generation process*

236 The cylinder pressure, heat release rate, and combustion characteristics of the
 237 optical engine fueled with FT, DF, PF, and BF are shown in **Fig. 4**. The prolongation of
 238 the ID varies with the type of oxygenated fuel blended into FT. Among them, the CNs
 239 of PODE₃ and FT are similar. Compared to FT, the PF with higher latent heat of
 240 vaporization exhibits a longer ID, and the combustion starting points are delayed.
 241 Additionally, due to the high latent heat of vaporization and lower heating value of

242 PODE₃, the peak cylinder pressure and PHRR of PF are relatively low. For n-butanol
 243 and DMC, the low CNs become the dominant factor that significantly prolongs the IDs
 244 and delays the combustion process for both BF and DF blended fuels. Especially, the
 245 lower viscosity and boiling point of DMC make a stronger vaporability, so the mixture
 246 formed is more homogeneous but more fuel-lean. DF requires a longer time to reach
 247 the concentration requirement of auto-ignition, thus the ID of DF is longer than that of
 248 BF. The longer IDs for BF and DF lead to higher premixed combustion ratios and result
 249 in shorter combustion durations (CDs). However, the ID of DF is too long to maintain
 250 desirable combustion timeliness compared to that of BF. The addition of n-butanol is
 251 more appropriate to improve the combustion process of FT.

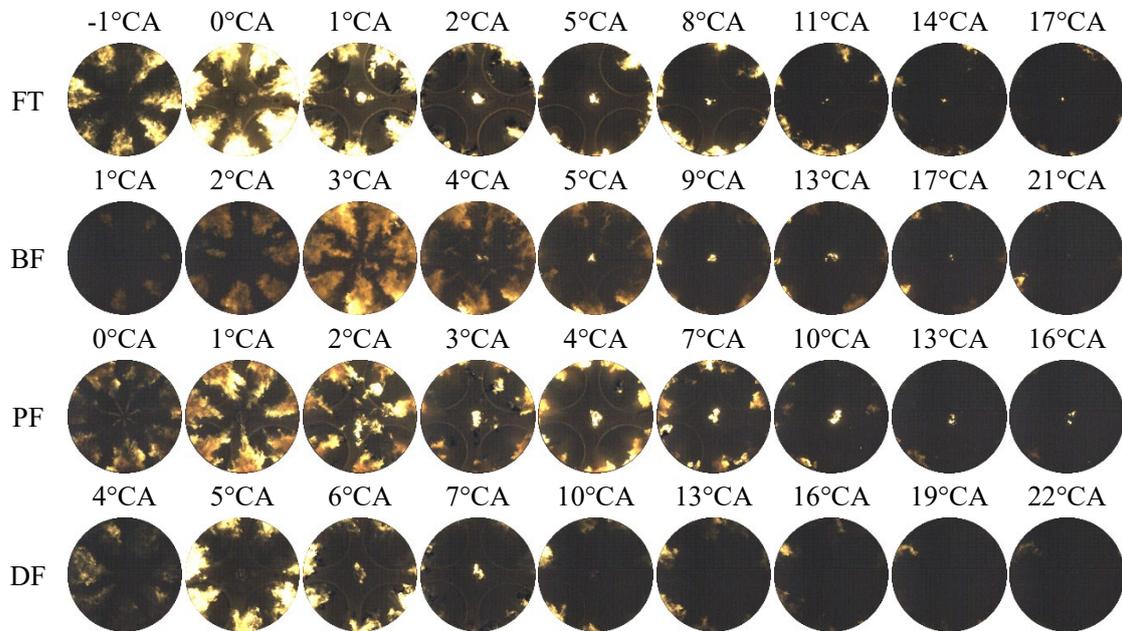


252 **Fig. 4.** Effects of oxygen-containing functional group structures on the FT synthetic
 253 diesel combustion process.

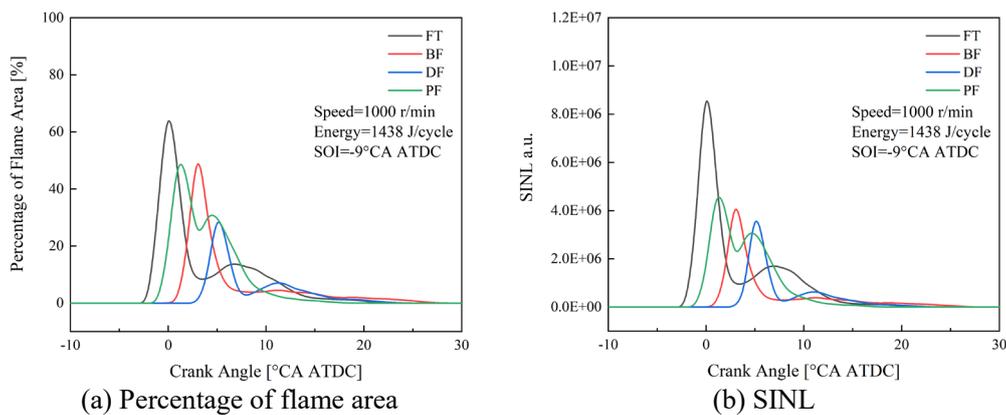
254 **Fig. 5** and **Fig. 6** show the effects of the structures of oxygen-containing functional
 255 groups on the history of flame development and characteristic parameters of FT. As it
 256 shows in the figure, a large area of spray-shaped bright diffusion flame can be seen
 257 during the combustion process of FT because of the high CN and short ID of FT. Such
 258 a pattern of combustion makes air-fuel mixing and combustion processes highly

259 overlapped and causes massive fuel cracking reaction under high-temperature and fuel-
260 rich conditions [72]. Therefore, the soot generation is significantly strengthened and
261 manifests as an extensive bright diffusion flame. During the intermediate stage of
262 combustion, the flame distribution is then concentrated towards the cylinder wall as a
263 result of fuel impingement. In the late stage of combustion, a punctiform flame appears
264 around the cylinder axis and exists for a long time. It is due to the needle valve seats at
265 the end of the injection process, resulting in the reduction of the injection pressure and
266 the spray penetration, leading to poor atomization. Additionally, due to the weak airflow
267 near the cylinder axis, diffusion combustion is conducted slowly by the poorly atomized
268 fuel here. After blending oxygenated fuels, the peaks of flame area percentage and SINL
269 are reduced significantly (FT>PF \approx BF>DF), and the flame appears in the sequence of
270 FT, PF, BF, DF. The self-carried oxygen of fuel can relieve the local fuel-rich
271 phenomenon caused by the non-uniform distribution of air-fuel mixture, inhibit the
272 generation of soot precursors, and promote the later oxidation of soot. Accordingly, the
273 generation of soot is suppressed, and the flame area and luminosity of oxygenated
274 blended fuels can be reduced. On the other hand, the introduction of oxygenated fuels
275 could adjust the physicochemical characteristics of the blended fuels, and promote the
276 air-fuel mixing indirectly by extending the ID and enhancing the vaporability. It can
277 improve the premixed combustion rate, and alleviate the problem of a large proportion
278 of diffusion combustion caused by high CN and relatively insufficient vaporability of
279 FT. Similar to FT, the flames of PF and BF are spray-shaped as well, but the bright
280 yellow area of PF flame is significantly smaller than FT, while BF flame shows dark

281 yellow overall. For DF with the longest ID, due to the highest premixed combustion
 282 rate and the homogeneity of combustion, the flame is mainly concentrated along the
 283 cylinder wall without showing the shape of the fuel spray explicitly.



284 **Fig. 5.** Effects of oxygen-containing functional group structures on the history of
 285 flame development.

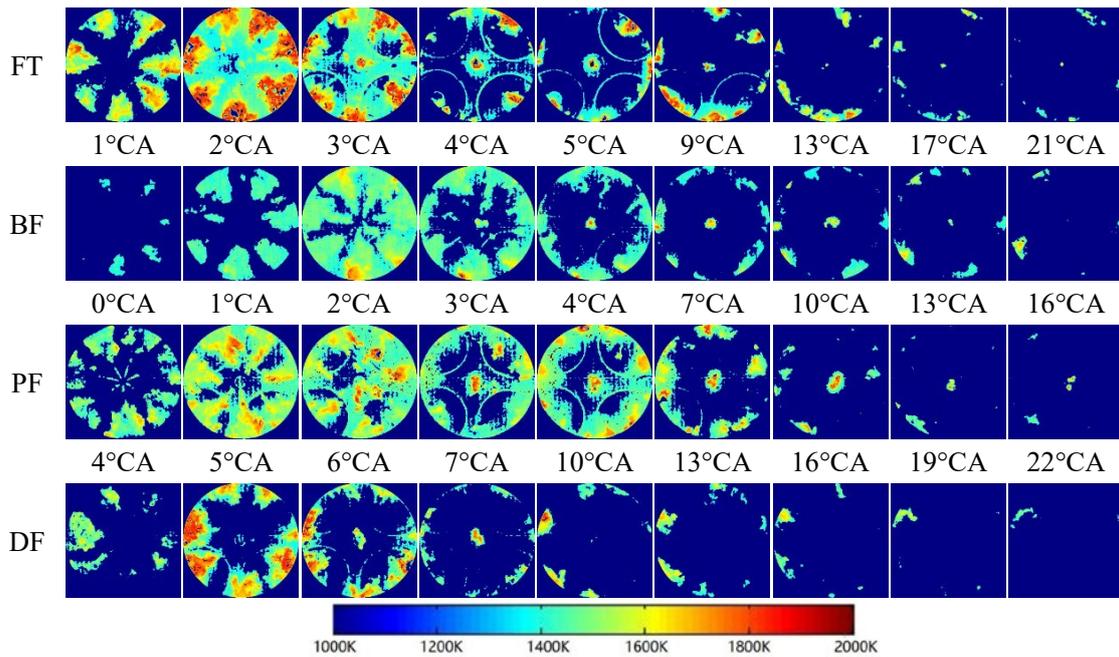


286 **Fig. 6.** Effects of oxygen-containing functional group structures on flame area and
 287 luminosity.

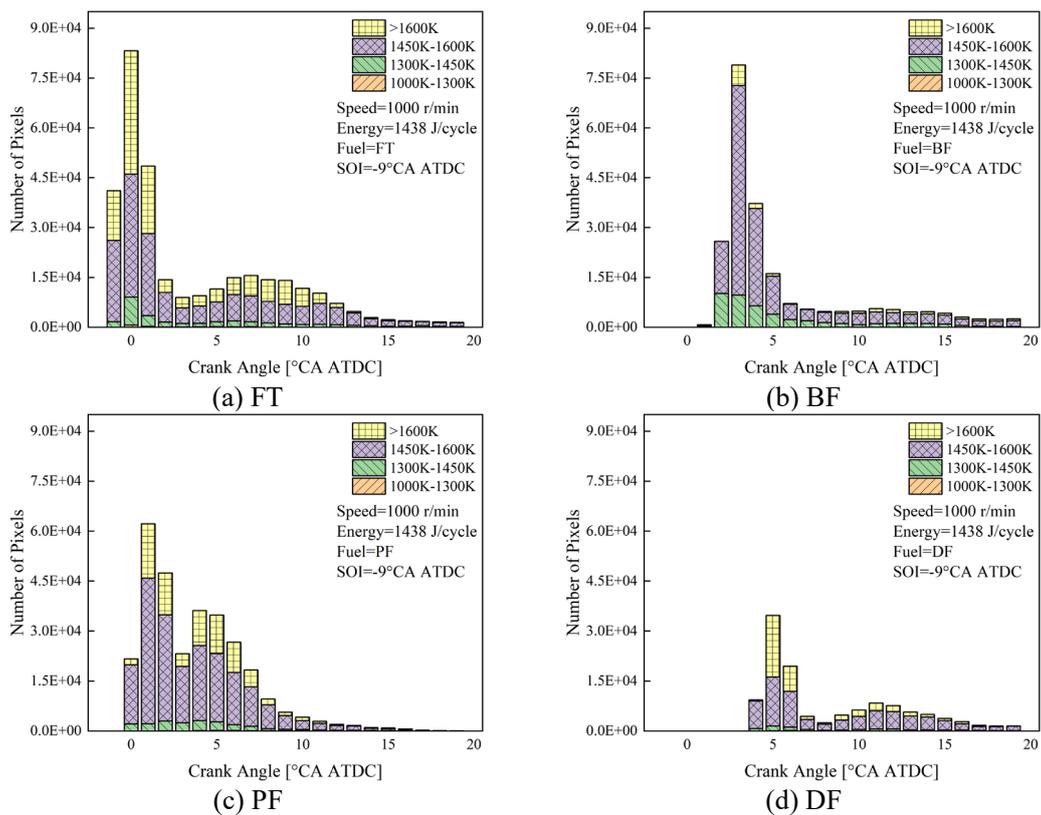
288 **Fig. 7** and **Fig. 8** show the distributions of in-cylinder temperature fields of the four
 289 fuels. Generally, the addition of oxygenated fuel results in the mitigation of high-
 290 temperature combustion phenomenon, showing up as the reductions in the area of the

291 local high-temperature and the number of high-temperature pixels especially the ones >
292 1600 K. Studies have pointed out that the main temperature range for soot formation is
293 1600 K-2600 K [73,74], so in order to balance thermal efficiency and soot emission
294 control, it is necessary to minimize the area of high temperature beyond 1600 K and
295 increase the average temperature of the area below 1600 K simultaneously, which is
296 equivalent to increasing the temperature of the heat source in the Carnot cycle. Among
297 the oxygenated blends, BF is the best in maintaining high thermal efficiency and
298 avoiding the soot generation temperature range simultaneously. The overall in-cylinder
299 temperature of the PF combustion process is lower than those of FT and BF, and the
300 number of high-temperature pixels above 1600 K is more than that of BF. In the cloud
301 map of temperature distribution, a large yellow and red region is present for PF, and the
302 duration of high-temperature area presence is also prolonged, representing a more
303 significant local high-temperature combustion phenomenon, which is not conducive to
304 the control of soot generation. In contrast, the temperature cloud map of the BF shows
305 a larger blue and green region and a smaller region in red, suggesting that the
306 temperature distribution during BF combustion is more uniform. For DF, due to the
307 delayed heat release process, the lower in-cylinder temperature and pressure caused by
308 the down going piston resulting in a lower heat release rate. Therefore, the overall in-
309 cylinder temperature of DF is lower than those of the other fuels, and the high-
310 temperature pixels (> 1600 K) are numerous and mainly distributed along the cylinder
311 wall.

-1°C CA 0°C CA 1°C CA 2°C CA 5°C CA 8°C CA 11°C CA 14°C CA 17°C CA



312 **Fig. 7.** The cloud maps of temperature distribution for blend fuels with different
 313 molecular structures.

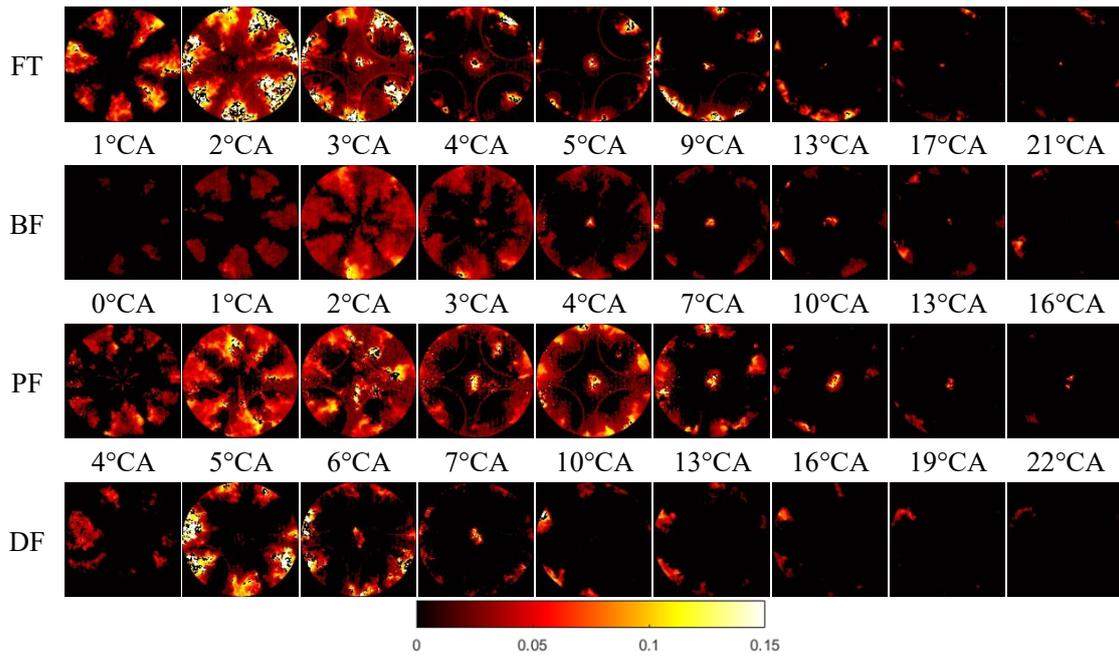


314 **Fig. 8.** The cumulative histograms of temperature distribution for blend fuels with
 315 different molecular structures.

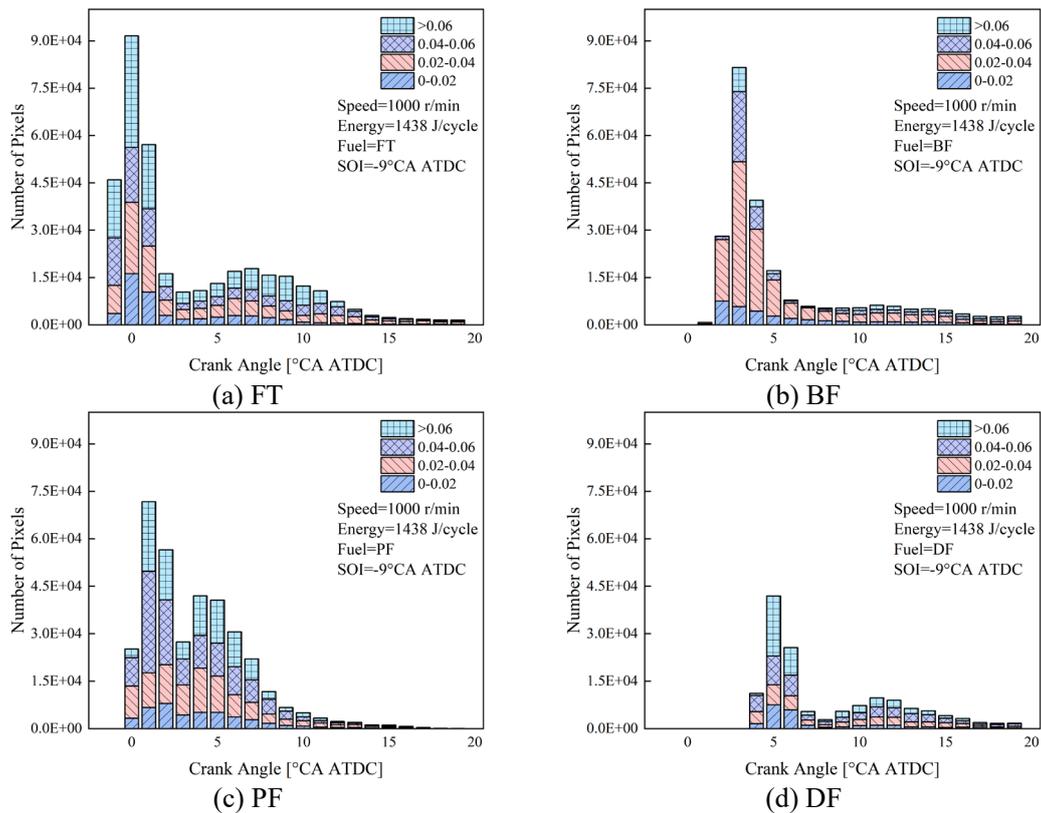
316 The KL factor field distributions of the fuels are shown in **Fig. 9** and **Fig. 10**. It can

317 be seen that the addition of oxygenated fuel can reduce the KL factor during the
318 combustion process to varying degrees. The KL factor field is more uniformly
319 distributed during the combustion process of BF, and the number of pixels with KL
320 factor > 0.06 is consistently lower compared to those of the other two oxygenated
321 blends. The KL factor of the BF reaches the peak at 3°CA ATDC and the peak is higher
322 than the peaks of the other two oxygenated blends, but the KL factor decreases rapidly
323 as the combustion proceeds. Although the peak KL factor of PF is lower than that of FT
324 and BF, the duration of KL factor presence is prolonged, the KL factor decreases slowly
325 during PF combustion process. In addition, there is an area of high concentration of
326 soot at the edges of PF flame, which is reflected in the cumulative histogram with a
327 higher number of pixels with KL factor > 0.06 . The main limiting factor for the soot
328 reduction capability of PODE₃ is the excessively high CN which leads to a short ID, an
329 overproportion of diffusion combustion, insufficient air-fuel mixing and accelerated
330 high-temperature fuel-rich cracking reactions. As the combustion process of DF is
331 postponed, the lower in-cylinder pressure and temperature could inhibit the generation
332 of soot, so the overall KL factor of DF is lower. However, it should be noted that the
333 combustion process of DF also suffers from the regions of high soot concentration, and
334 the variation of the KL factor shows a double-peaked tendency which is similar to those
335 of FT and PF. In summary, among the three oxygenated blends, BF has the strongest
336 ability in inhibiting soot generation, which is consistent with the findings of other
337 studies ^[75-77].

-1°CA 0°CA 1°CA 2°CA 5°CA 8°CA 11°CA 14°CA 17°CA



338 **Fig. 9.** The cloud maps of soot KL factor distribution for blended fuels with different
 339 molecular structures.



340 **Fig. 10.** The cumulative histograms of soot KL factor distribution for blended fuels
 341 with different molecular structures.

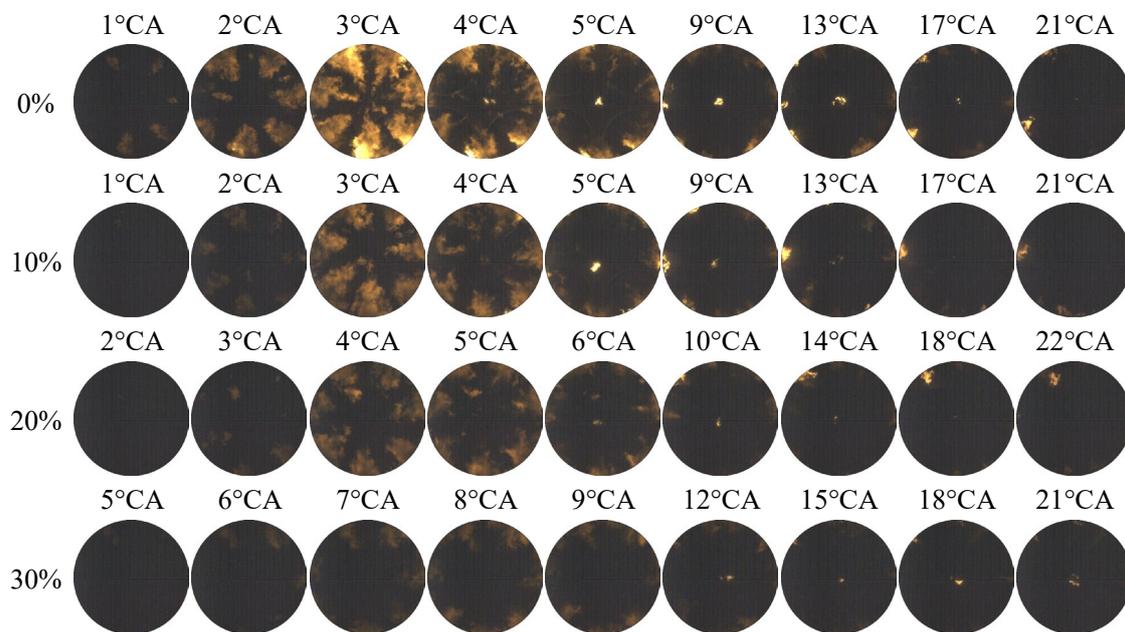
342 *3.2 Effects of internal and external EGR on combustion and soot generation of n-*

343 *butanol/FT synthetic diesel blend*

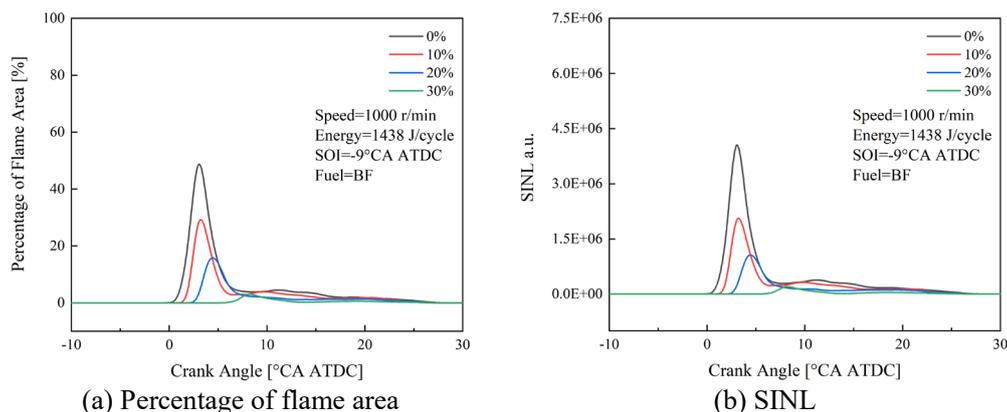
344 Among the three oxygenated fuels used in this study, n-butanol has the outstanding
345 ability in the control of soot generation, and the addition of n-butanol to FT could
346 maintain better combustion timeliness without prolonging the CD excessively, which
347 ensures an acceptable fuel economy together with restrained soot emission. Therefore,
348 BF is selected in this section as the test fuel to investigate the effects of different EGR
349 introducing schemes and EGR rates on the combustion process.

350 **Fig. 11** and **Fig. 12** show the effects of different ratios of external EGR on the flame
351 development history and flame characteristics of BF. It can be noticed that the external
352 EGR presents an inhibitory effect on BF combustion. As the external EGR rate
353 increases, the percentage of diffusion flame area and the SINL decrease, the ID is
354 prolonged, and the timing of flame appearance is significantly delayed. The cylinder
355 temperature before combustion is decreased because of the higher specific heat of in-
356 cylinder gas, and the reactivity of the air-fuel mixture is reduced by the decline of
357 oxygen concentration in the cylinder ^[78,79], the two factors above lead to the
358 prolongation of the ID and the retardation of flame appearance. The prolonged ID is
359 conducive to improving the uniformity of the air-fuel mixture, resulting in a lower
360 diffusion combustion rate and reducing the concentration and temperature gradients of
361 the combustion process. The cracking reactions of fuels are less likely to take place,
362 leading to reductions in soot generation, diffusion flame area and luminosity. The
363 dilution and the chemical equilibrium closer to reactants caused by external EGR lead
364 to a reduction of the combustion rate, which could also reduce the in-cylinder

365 temperature and inhibit the generation of soot. According to **Fig.11**, as the external EGR
 366 rate increases, the flame luminosity along the cylinder wall decreases. This is caused
 367 by the strong airflow near the cylinder wall, which helps the mutual diffusion and air-
 368 fuel mixing. The fuel around the cylinder axis with insufficient injection pressure and
 369 airflow intensity suffers from poor atomization and a slow mixing rate, so the flame
 370 near the cylinder axis does not fade away until the EGR rate reaches 20%.



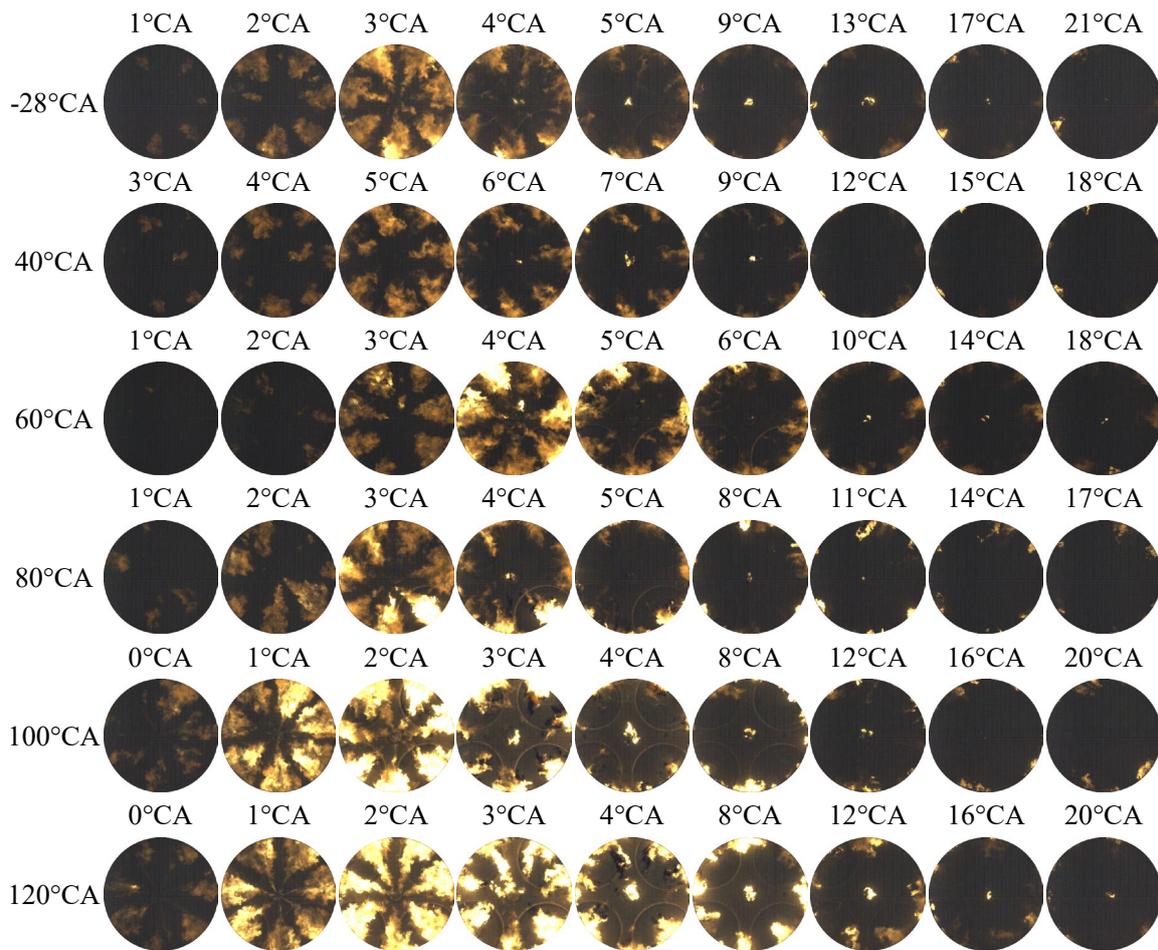
371 **Fig. 11.** Effects of external EGR rates on the history of BF flame development.



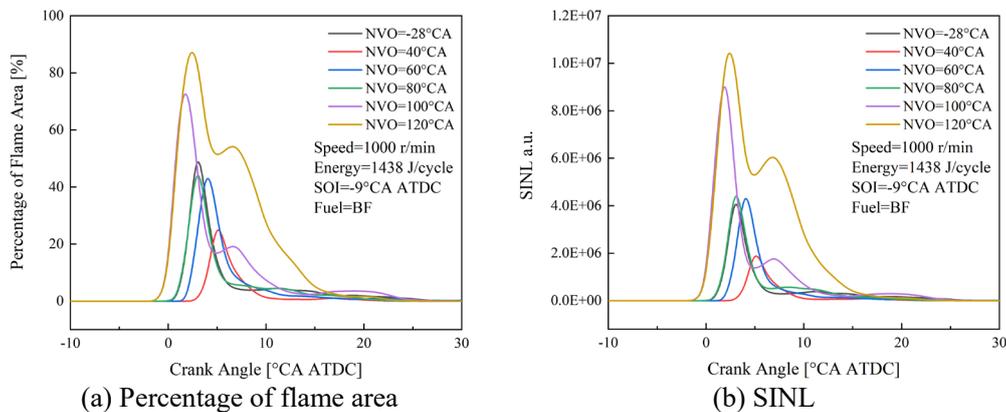
372 **Fig. 12.** Effects of external EGR rates on BF flame area and luminosity.

373 **Fig. 13** and **Fig. 14** show the BF flame development and flame characteristics
 374 under different internal EGR rates. Unlike the external EGR, it shows an effect of

375 inhibition first and then promotion on the flame area and luminosity as the internal EGR
376 rate increases. When the internal EGR rate is low (NVOA=40°CA), the flame area and
377 luminosity are significantly lower compared with those at the original valve timing
378 (NVOA=-28°CA), and the timing of flame appearance is significantly delayed. As the
379 NVOA reaches 60°CA, the flame area, SINL and the timing of the flame appearance
380 are similar to those of the original valve timing. As the internal EGR rate is increased
381 further, a significant promotion of the ignition process is shown. When
382 NVOA=120°CA, the ID is shorter than that of the original valve timing obviously,
383 resulting in insufficient air-fuel mixing. These changes are very unfavorable to the
384 control of soot emission, therefore making the flame area percentage and SINL almost
385 twice that of the original valve timing. Li et al. suggested that internal EGR could affect
386 the combustion process in two ways: the dilution effect of exhaust gas with higher
387 specific heat, and in contrast, the heating effect of high-temperature residual gas ^[80].
388 Under the lower rates of internal EGR, less exhaust gas is trapped and the heating effect
389 on the in-cylinder temperature is weaker, the inhibition effect takes the leading role. As
390 the internal EGR rate increases, the heating effect increases significantly, and the
391 promotion effect becomes dominant gradually. In addition, the implementation of
392 internal EGR requires an alteration of valve timing away from its optimal value,
393 resulting in decreases in the volumetric efficiency and the amount of fresh air, so the
394 heating effect is additionally enhanced. Accordingly, to restrain the in-cylinder
395 temperature using internal EGR, an optimal internal EGR rate is supposed to be found,
396 and an excessive internal EGR rate will lead to further deterioration of combustion.



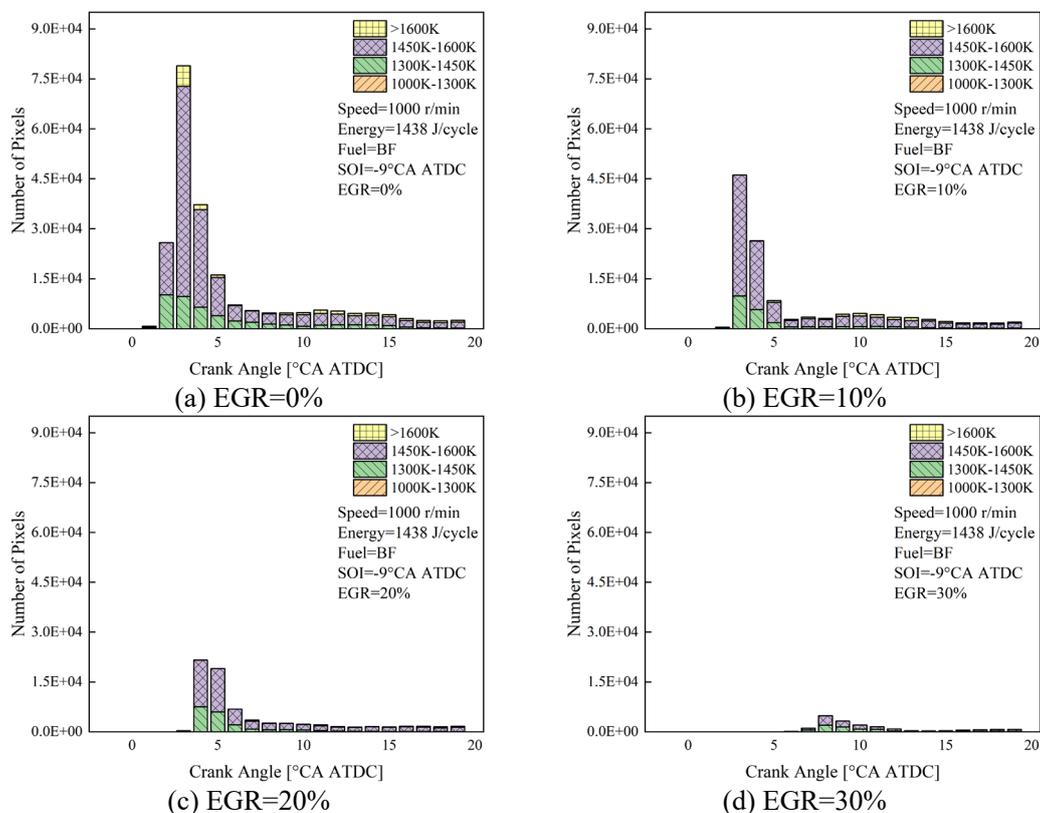
397 **Fig. 13.** Effects of internal EGR rates on the history of BF flame development.



398 **Fig. 14.** Effects of internal EGR rates on BF flame area and luminosity.

399 **Fig. 15** shows the distributions of the in-cylinder temperature field of BF at
 400 different external EGR rates. As shown in the figure, with the increase of external EGR
 401 rate, the total number of high-temperature pixels decreases generally, the pixels with
 402 temperatures > 1600 K almost disappear, and the timing of high-temperature region

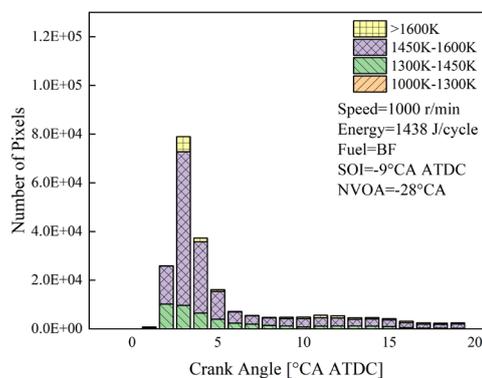
403 appearance is delayed. External EGR could effectively reduce the proportion of
 404 diffusion combustion while suppressing the intensity of the combustion process, thus
 405 avoiding local fuel-rich combustion and the temperature range in which soot is
 406 generated significantly, providing favorable conditions for the control of soot emission.
 407 However, under high-ratio external EGRs, the in-cylinder temperature is suppressed
 408 significantly, and the combustion process is delayed excessively, which negatively
 409 affects the economy obviously.



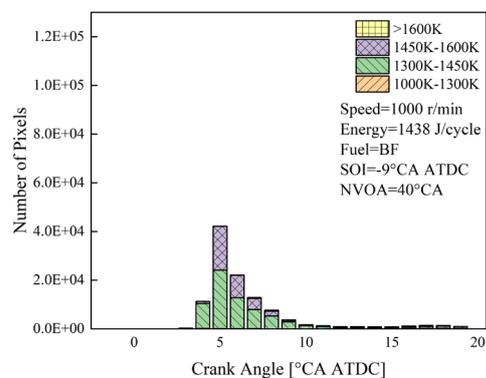
410 **Fig. 15.** The cumulative histograms of temperature distribution for different external
 411 EGR rates.

412 The distributions of the in-cylinder temperature field of BF under different internal
 413 EGR rates are shown in **Fig. 16**. Since the lower internal EGR rates could prolong the
 414 ID appropriately and provide a sufficient air-fuel mixing process, the gradients of in-

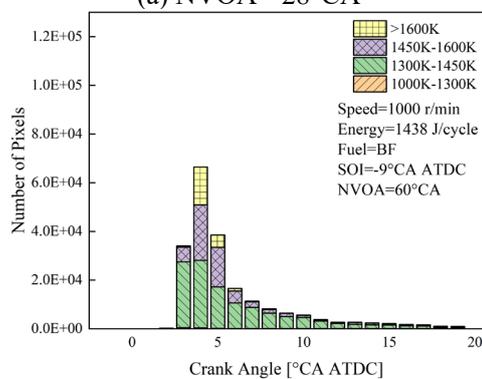
415 cylinder concentration and temperature are reduced, resulting in fewer high-
 416 temperature pixels when NVOA=40°CA. As the internal EGR rate increases, the
 417 promotion effect of internal EGR on in-cylinder temperature gradually dominates.
 418 When the NVOA exceeds 80°CA, the high-temperature combustion phenomenon is
 419 more obvious compared to the original valve timing, and the long-lasting high-
 420 temperature pixels during the combustion process indicate that the combustion process
 421 is prolonged overly. Meanwhile, the excessive heat transfer between the gas and the
 422 cylinder wall leads to a decrease in thermal efficiency. Additionally, the over-
 423 proportioning of diffusion combustion caused by a high ratio of internal EGR leads to
 424 significant increases in the number of high-temperature pixels above 1600 K, which do
 425 not contribute to the control of soot emission.



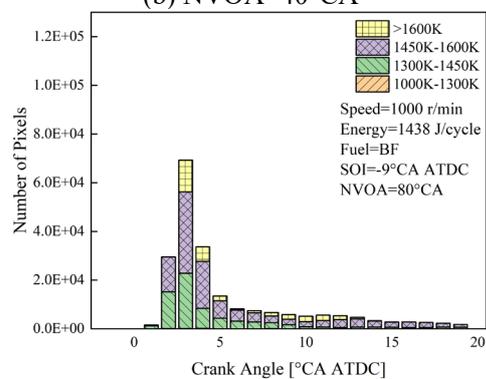
(a) NVOA=-28°CA



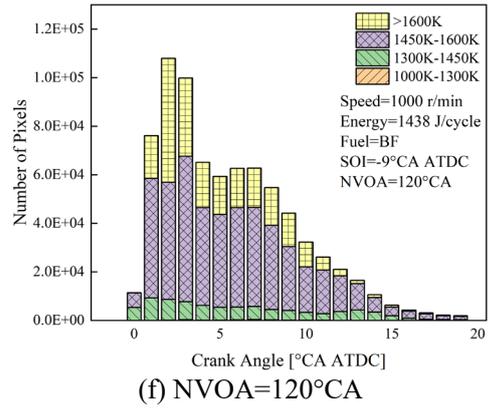
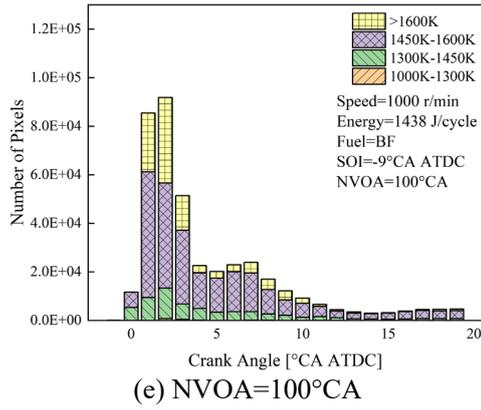
(b) NVOA=40°CA



(c) NVOA=60°CA

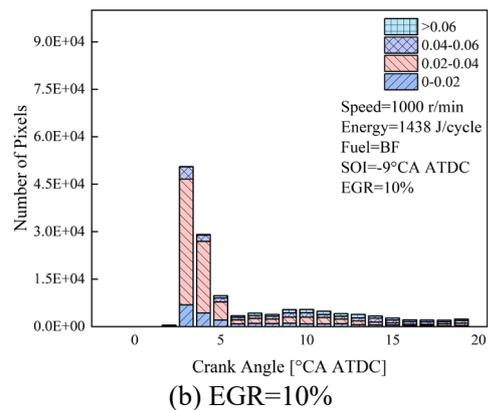
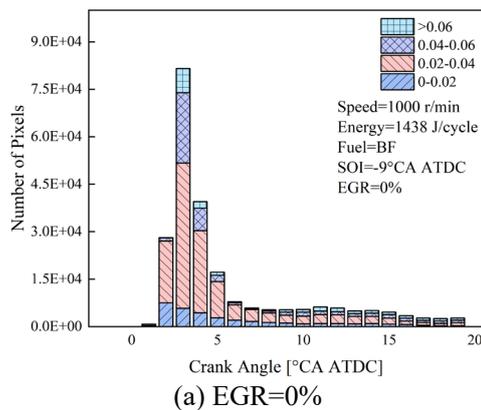


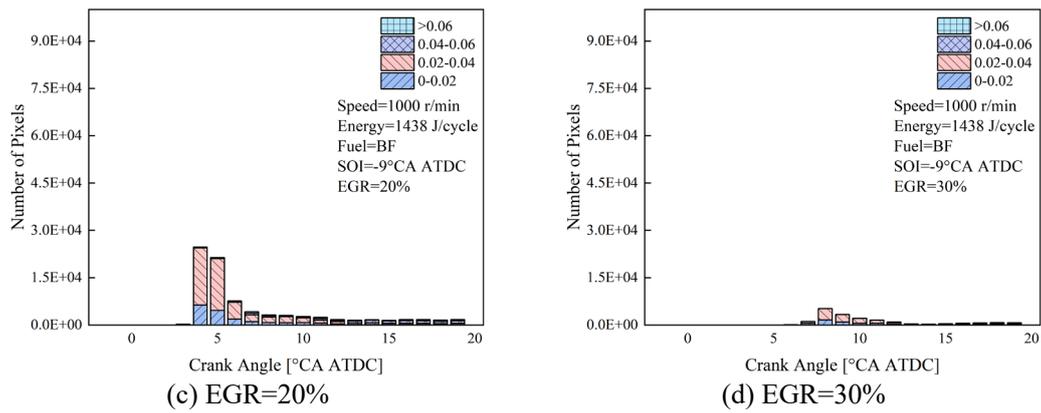
(d) NVOA=80°CA



426 **Fig. 16.** The cumulative histograms of temperature distribution for different internal
 427 EGR rates.

428 The effect of external EGR on the KL factor field distribution of BF fuel is shown
 429 in **Fig. 17.** Under the combined effect of external EGR and the addition of n-butanol,
 430 the improved air-fuel mixing process leads to a lower diffusive combustion rate as well
 431 as lower gradients of in-cylinder temperature and concentration. Therefore, the local
 432 high-temperature fuel-rich combustion problem is alleviated, resulting in a significant
 433 reduction of KL factor. Although the soot KL factor is reduced significantly with a high
 434 ratio of external EGR, the delayed timing and the lowered amount of soot generation
 435 indicate a reduced thermal efficiency.





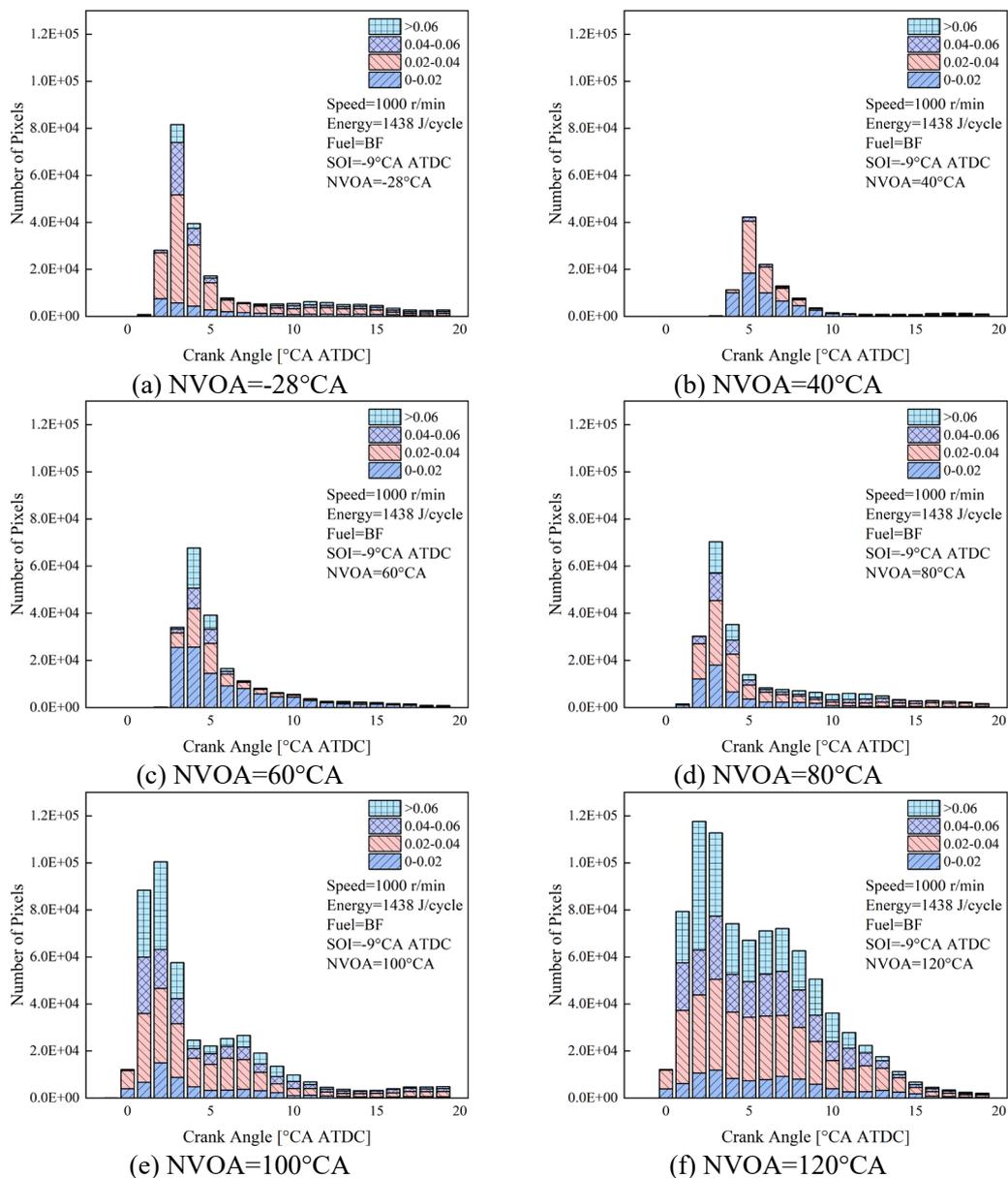
436 **Fig. 17.** The cumulative histograms of soot KL factor distribution for different
 437 external EGR rates.

438 The effect of internal EGR on the KL factor field distribution of BF fuel is shown
 439 in **Fig. 18**. Since the low rate of internal EGR could effectively restrain the high-
 440 temperature combustion and break the conditions of soot generation, the KL factor
 441 decreases significantly and the timing of soot generation is postponed remarkably. As
 442 the internal EGR rate increases, the inhibitory effect is gradually transformed into a
 443 facilitating effect, the insufficient air-fuel mixing leads to an increased KL factor and
 444 the advance of soot generation timing. When the NVOA > 80°CA, as a result of
 445 deteriorated combustion and excessive in-cylinder concentration and temperature
 446 gradients, the pre-inhibition and later oxidation effects of internal EGR on soot are
 447 weakened, resulting in a significantly higher KL factor compared to the ones with the
 448 original valve timing. According to the previous simulation study by our group, the
 449 amounts of trapped residual gas at the NVOAs of 60°CA and 100°CA are
 450 approximately the same as the amounts of exhaust gas introduced under 10% and 20%
 451 external EGRs, respectively. Under the conditions of 10% external EGR and
 452 NVOA=60°CA, it shows that the inhibiting effect of internal EGR is weaker than the

453 effect of external EGR at the current condition. Under 20% external EGR and
454 NVOA=100°CA operating conditions, the external EGR maintains a strong inhibitory
455 effect, while the promotion effect exhibited by the internal EGR is rather more
456 significant than its inhibitory effect. The following factors are responsible for this
457 difference. Firstly, in this study, the trapped high-temperature gas has a stronger heating
458 effect than the gas introduced by external EGR, especially under conditions of high
459 EGR ratios. Secondly, the trapped gas has a lower specific heat because of the lower
460 proportion of triatomic gas, and it has a certain reaction activity, leading to a weaker
461 inhibitory effect. The significant impact of the temperature of the EGR gas on the
462 combustion process was also emphasized in reference [79]. Finally, internal EGR leads
463 to an incomplete and insufficient gas exchange process and a reduction of in-cylinder
464 charge [46,81], which further aggravates the heating effect of residual gas and local fuel-
465 rich phenomenon. In contrast, with external EGR, due to the low engine load and
466 relatively high ϕ_a in this test, the in-cylinder oxygen is sufficient to inhibit soot
467 generation. It must be noted that both internal and external EGR cause severe
468 deterioration of the combustion process when a high EGR rate is adopted. The large
469 proportion of external EGR causes excessively low in-cylinder temperatures and
470 delayed heat release processes, resulting in poor combustion timeliness and stability.
471 The large proportion of internal EGR leads to advanced ignition timing and insufficient
472 air-fuel mixing significantly, which also causes unstable combustion and extensive soot
473 generation. In addition, the negative impact of the introduction of EGR on energy
474 availability deserves attention, as confirmed by the previous exergy assessment [82].

475 Therefore, in practice, both the internal and external EGR rates have to be controlled

476 strictly.



477 **Fig. 18.** The cumulative histograms of soot KL factor distribution for different
478 internal EGR rates.

479 4 Conclusions

480 In this study, an optical diagnostic study was conducted on the combustion and soot
481 generation history of FT synthetic diesel and its oxygenated blends under different EGR
482 introducing schemes based on a self-built optical engine. The findings of the study can

483 be summarized as follows:

484 1. The additions of n-butanol, POE₃, and DMC could directly inhibit generation
485 and promote later oxidation of soot via the oxygen-carrying properties to varying
486 degrees, and adjust the physicochemical properties of blended fuels in various ways to
487 promote air-fuel mixing and thus control the generation of soot. Blending oxygenated
488 fuels with FT synthetic diesel could reduce the in-cylinder flame area and luminosity.
489 For oxygenated blends, the local high-temperature fuel-rich combustion phenomenon
490 is alleviated in different degrees, and the KL factors of soot are reduced. Among the
491 three oxygenated fuels, n-butanol exhibits the strongest ability for soot reduction,
492 followed by POE₃ and DMC.

493 2. The external EGR shows an inhibitory effect on the ignition and combustion
494 processes. As the external EGR rate increases, the combustion rate is decreased, the
495 heat release process of combustion is delayed, the in-cylinder flame area and luminosity
496 are reduced, and the in-cylinder temperature and KL factor are lowered. Because of the
497 minor effect of external EGR on the gas exchange process and the low engine load, the
498 phenomenon of local fuel-rich combustion under a high ratio of external EGR is not
499 obvious in this test.

500 3. As the internal EGR rate increases, it shows the effect of inhibiting first and then
501 promoting on the ignition and combustion processes. Low-ratio internal EGR could
502 reduce the reaction rate, diffusion flame area and luminosity, as well as the in-cylinder
503 temperature and KL factor. The heating effect of the high-ratio internal EGR shortens
504 the ID significantly, resulting in insufficient air-fuel mixing and obvious increases in

505 the in-cylinder temperature and KL factor.

506 4. For EGR schemes with the same introduced amounts, the pure CO₂ from external
507 EGR has a stronger inhibiting effect on the heat release process than the effect of
508 residual gas trapped by internal EGR. In practice, both high ratios of internal and
509 external EGR lead to an excessive deterioration of combustion and a decrease in
510 combustion stability and therefore need to be strictly controlled. The strategy of
511 properly proportioned internal/external EGR with oxygenated fuel additives could
512 effectively optimize the combustion process of FT synthetic diesel and expand the range
513 of high-efficiency-clean-combustion.

514 **Notes**

515 The authors declare no competing financial interest.

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