Optical Soot Measurement of Bio-Butanol Upstream Product, ABE (Acetone-Butanol-Ethanol), under Diesel-like Conditions

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Abstract:

ABE (Acetone-Butanol-Ethanol), the upstream product of bio-butanol, is considered as a promising alternative fuel for IC engine. Two-dimensional soot distribution and soot mass of spray combustion of ABE and diesel was measured by forward illumination light extinction (FILE) technology. The ambient temperature and oxygen concentration of injection was varied from 1200K to 900K and 21% to 11% to represent normal temperature combustion and low temperature combustion modes respectively. It was found that soot distribution area and intensity of ABE, especially ABE with a high acetone fraction, is much lower than that of diesel. The soot clouds area and intensity increase with increasing ambient temperature and decreasing oxygen concentration, but the increased scale of soot for ABE is much lower than that of diesel. Quantitative results of soot mass for ABE show that the soot increases rapidly with increasing ambient temperature at lower oxygen concentration conditions because the soot producing rate is largely increased when elevating temperature, while the soot oxidation rate remained slow due to the insufficient oxygen. Based on analysis, it was

found that clean combustion of ABE benefited from a lean air-fuel mixture because of the great volatility and low stoichiometric ratio of the fuel, and the competition of Carbon atom between CO and C₂H₂ in the pyrolysis reactions that reduced the soot source materials. Overall, as an alternative IC fuel, ABE is able to achieve much lower soot than diesel and is able to maintain clean combustion in a large range of environmental conditions.

Keywords: ABE (Acetone-butanol-ethanol), bio-butanol, spray combustion, Soot measurement, forward illumination light extinction, diesel-like conditions

1 Introduction

Along with the development of the auto industry, transportation promoted the development of human society and brought many conveniences to modern life. However, the rapid growth of vehicles also brought worldwide problems, such as energy crisis and air pollution. In China, the oil consumed by transportation accounted for 49.6% of that in the whole country, which is almost equal to the net oil import [1]. At the same time, air pollution is becoming a serious problem all over the world, especially for the developing countries, such as India and China. The concentration of hazardous particles in Beijing, China, in January 2013, was forty times the level deemed safe by the World Health Organization. Additionally, based on a 2012 Asian Development Bank report, less than 1 percent of China's 500 largest cities meet the WHO's air quality standards [2].

45 In order to mitigate the problems of the energy crisis and air pollution, it is
46 imperative to develop clean and highly efficient combustion systems in the
47 transportation sector. It is known that the thermal efficiency of diesel engine, operated
48 by compression ignition (CI), is much higher than that of gasoline engine, operated by
49 spark ignition (SI). The modern 'clean' automotive diesel engine is 20-40% more fuel
50 efficient than its gasoline counterpart [3]. Therefore, the application of CI diesel engines
51 itself is an effective way for energy saving. Beside the high efficiency, the application of
52 the diesel engine is also beneficial due to its high output power, cost effectiveness etc.
53 This is the reason why European countries promote the diesel engine in not only
54 commercial vehicles, but also in cars [4]. However, high level smoke emissions is one of
55 the primary concerns for diesel engines. In China, cities do not permit commercial
56 vehicles to enter urban areas due to its heavy smoke emissions [5]. Besides the
57 combustion system, development of alternative fuels, especially renewable fuels, is
58 another potential way to mitigate the energy crisis [6]. In last few decades, alcohols,
59 biodiesels, DME, natural gas, LPG, and hydrogen etc. have been proposed as alternative
60 transportation fuels and explored [7,8,9,10,11]. The alcohols were regarded as some of
61 the best potential alternative IC engine fuels due to its excellent combustion
62 performance and renewable characteristics [12]. As a result, the combination of a diesel
63 combustion model and renewable alcohols is a potential choice to take the full
64 advantages from both the combustion system side and the fuel side. These researched
65 works actually have proven the feasibility of this attempt [13,14,15,16]. It was found
66 that with the addition of methanol or ethanol, the engine-out HC and CO emissions

could be reduced slightly while PM emissions could be reduced largely. At the same time the thermal efficiency also can be increased slightly by adjusting the combustion phasing due to the rapid combustion.

Among alcohols, butanol has been recognized as a superior next-generation biofuel with commercialization potential due to its excellent fuel properties. Compared to short-chain alcohols such as ethanol and methanol, butanol has properties similar to traditional transportation fuels due to the longer hydrocarbon chain and has the capability to work with vehicles designed for traditional fuels without any modifications [17,18,19]. Butanol also has a higher energy density than ethanol, which leads to reduced power and can also be used either in neat form or in blends with fossil fuels. It has a relatively high cetane number and excellent miscibility with diesel, thus making it suitable to be an alternative fuel for diesel. Also, n-butanol is much less hygroscopic and corrosive, and therefore can be considered as a “drop-in” fuel that would be compatible with the current fuel distribution infrastructure [16].

Even though bio-butanol has been regarded as one of the potential alternative transportation fuels, the extraction of bio-butanol from ABE fermentation, the most common method to produce pure butanol, is faced with a high separation and purification cost from the fermentation broth. Separating butanol from the broth by traditional methods, i.e., direct distillation, requires 18.4 MJ/kg, which is approximately 54% of its combustion heat value [20]. If the upstream product of bio-butanol, the acetone-butanol-ethanol mixture (ABE), could be used for clean combustion directly in IC engines, the separation costs would be mitigated.

89 Researchers of ABE fermentation first realized the potential of ABE used as
90 alternative fuel for IC engines [21,22]. It was found that by using biodiesel as extractant
91 during the fermentation process, the ABE-biodiesel solution was obtained directly and
92 at the same time the productivity of ABE also increased. Chang et al. [23] first reported
93 the application of ABE fuel on internal combustion engines in ABE-diesel blends mode.
94 They found that with the 20% ABE addition, there was a reduction of both particulate
95 matter (PM) and total toxicity equivalency of polycyclic aromatic hydrocarbons by
96 approximately 20%, but suffered an increase in nitrogen oxides (NOx). The diesel engine
97 also demonstrated an increase of brake thermal efficiency up to 8.56% in certain ABE-
98 diesel blends. In another work [24], they extended the application of ABE-diesel blends
99 to ABE-biodiesel-diesel blends. The results showed that the use of ABE-biodiesel-diesel
100 blends could simultaneously reduce both PM and NOx by 4.30-30.7% and 10.9-63.1%
101 respectively comparing with regular diesel. It eliminated the high NOx emission problem
102 for biodiesel applications on the diesel engine. These works primarily proved the
103 feasibility of ABE applications on diesel engines.

104 In order to clearly understand the spray combustion process of ABE-diesel blends in
105 a fundamental way, some further research has been conducted. Van Geem et al. [25]
106 built a detailed mechanism containing 350 species and more than 10,000 reactions for
107 the pyrolysis and oxidation of ABE based on a more complicated mechanism for
108 hydrocarbon fuels, which has good agreement with observed experimental data. Ma et
109 al [26], investigated the evaporation characteristics of ABE and diesel blend droplets
110 experimentally through droplet suspension techniques at different ambient

temperatures (423-823 K) under normal gravity and atmospheric pressure. The results show that the droplet lifetime of ABE is always shorter compared to that of diesel due to the higher volatility of the ABE components.

The author's group investigated the spray and combustion characteristics of ABE and its blends with diesel on an optical constant volume chamber with a high speed camera coupled with a copper laser sheet [27,28,29,30]. It was found that ABE has the similar spray and flame characteristics to neat n-butanol. That meant the upstream product of bio-butanol kept the capability to be an excellent alternative fuel for IC engines, but with a lower cost. Similar to the blends of butanol-diesel, with the addition of ABE, the blends displayed a delayed combustion phasing. But the delayed combustion phasing phenomenon can be eliminated in some degree by adjusting the ABE ratio or the ABE components ratios. By increasing the ratio of acetone in the ABE solution, the heat release rate of spray combustion is almost exactly the same as that of neat diesel.

More importantly, all of the works done in the author's group indicated that the natural flame luminosity during the spray combustion of ABE and ABE-diesel blends is significantly lower than that of neat diesel. The natural luminosity is contributed by two sources, which are chemiluminescence and soot incandescence, with the latter is much stronger than the former one. Thus, the soot reducing potential of the ABE addition was stated based on the phenomenon of natural flame luminosity [31].

However, the soot incandescence is sensitive to the local temperature of the burning region, which is possibly influenced, and typically reduced, by the high latent heat of the ABE components [32]. In order to determine the real soot reduction effect of

ABE during the spray combustion process, it is necessary to investigate the soot emission characteristics in a more direct way other than estimating by natural flame luminosity intensity. Based on the motivation mentioned above, the soot distribution and concentration of neat ABE spray combustion was measured and compared with that of neat diesel through forward illumination light extinction (FILE) under different ambient temperatures and oxygen concentrations in a constant volume chamber.

2 Experimental methodology

2.1 Test System

A test system based on a constant volume chamber with a bore of 110 mm and a height of 65 mm was used in this study. The high temperature/pressure environment is built by pre-combustion of oxygen, nitrogen and acetylene to mimic the cylinder condition of diesel engine near the top dead center. The chamber has an open end on the top with a fused silica window and a hydraulic-actuated electronic-controlled unit injector (HEUI) installed at the bottom allowing optical access (see Figure 1). The window has dimensions of 130 mm in diameter and 60 mm in thickness, with a high UV transmittance down to 190 nm. The cylinder wall is heated to 380K during the experiment to mimic the wall temperature of a diesel engine as well as to prevent water condensation on the optical window. The pre-mixture is ignited by a spark plug with a plasma coil. Pertinent information during combustion process was recorded by a high speed camera, Phantom V7.1, with a 105-mm focal length lens (Nikkor) located above the chamber. The camera is implanted with a SR-CMOS 12-bit 800×600 pixel

monochrome sensor with a spectrum response from 400 to 1000 nm. The light source is supplied by a copper vapor laser, Oxford Lasers LS20-50, which can be externally controlled and run up to a maximum frequency of 50 kHz with a pulse duration of 25 ns. The high speed camera is synchronized to the laser beam by a DG535. Depending on the various experiments , extra devices such as diffusers and filters might be included in the test system. The detailed experimental setup can be found in Refs [29].

2.2 Test Method and Data Processing

Soot distribution and concentration of spray combustion was measured by forward-illumination light-extinction method in this study, which was developed by Xu and Lee [33]. It is similar to the normal light-extinction method but the Light source and camera are placed at the same side of the flame, thus a light diffuser is needed, as shown in Figure 2. Due to the applied light diffuser, the incident light passes the soot cloud twice and is reflected by the diffuser once. Thus a modified form of the Lambert-Beer's law can be obtained based on the intensity of reflection light under conditions with and without soot cloud, as seen in equation (1).

$$I = I_0 \exp\left(-\int_0^{2L} K_{ext} dx\right) \quad (1)$$

Where I and I_0 are the reflected light intensities with and without of the soot cloud respectively, K_{ext} is the extinction coefficient for soot cloud, and L is the thickness of the soot cloud.

Since the thickness of the soot cloud is usually unknown, what is obtained from the experiment is the line-of-sight instead of the soot concentration. Based on the Rayleigh approximation, it can be expressed in equation (2).

$$C_v L = \frac{\lambda}{2K_a} \ln\left(\frac{I_0}{I}\right) \quad (2)$$

Where λ is the wavelength of the mono-color light and K_a is the dimensionless absorption constant which is selected as 4.9[34]. This value was obtained from the validation of Rayleigh approximation for soot measurements with primary soot sizes below 50 nm and laser wavelengths of 511 nm.

The total time-related soot mass can be obtained by summing all the local soot mass at each pixel, which can be expressed as

$$m = \sum \rho_s C_v L (\Delta r_i)^2 \quad (3)$$

In equation (3), it is assumed that the area represented by each pixel is a square with dimension of Δr_i , and the mean soot mass density ρ_s is 2.0 g/cm³ [35], and i is the index of pixel.

In addition, a few images of natural flame luminosity or CH* chemiluminescence are also involved in the discussion section. The natural luminosity images are recorded by high speed camera without a laser beam and a light diffuser. CH* chemiluminescence images are recorded by the system setting similar to that of natural luminosity settings but with a band-pass filter, longer exposure, and greater aperture.

2.3 Fuel Preparations

An ABE solution is prepared by mixing analytical grade acetone (99.5%), butanol (99.5%) and ethanol (99.8%), rather than obtaining the mixture through ABE fermentation. The solution with a volumetric fraction of 3:6:1 (acetone: butanol: ethanol), which is the typical composition of the product of ABE fermentation, is named as ABE(3:6:1), in this paper, or as ABE. In addition, ABE component ratios can be controlled during the ABE fermentation process with the development of fermentation technology [36,37]. Thus a higher acetone containing ABE solution with a volumetric ratio of 6:3:1 (acetone: butanol: ethanol), named as ABE(6:3:1) is also studied. Diesel is selected as the base fuel in this study. The properties of individual fuels and blends are listed in Table 1.

Since the measure region is located in the combustion area, which is in the downstream of lift-off length, the droplet cloud could not influence the soot measurement because the maximum penetration distance of any liquid-phase fuel, is approximately coincident with the lift-off length [34]. It can be also inferred that the accuracy of FILE measuring is insensitive to injected fuel types.

2.4 Test Conditions

The test was started by filling the chamber with the premixed mixture which included acetylene, oxygen, and nitrogen. The mixture was then ignited by a spark to create a high-temperature/pressure environment inside the chamber, but slowly cools down due to the heat loss. The spray injection and camera was triggered simultaneously

when the pressure reached the desired test conditions. Temperature, density, and oxygen concentrations of the gas at the time of injection in the chamber could be controlled as desired by adjusting injection timing and partial pressure[27]. However, the ambient density was kept constant at 14.8 kg/m^3 in this study, which is the typical cylinder condition at top dead center (TDC) for diesel engine. The test conditions were summarized in table 2.

It is noted that images were captured in 3-5 injections for each tested conditions, and the individual images are averaged before used in result discussion. Three Individual as well as mean images for one of the tested cases are shown in Fig. 3 in order to confirmed the repeatability similar to that in ref. [41]. Individual images from the different injections show the similar soot distributions. Since the images are mainly used for qualitative comparison, the scales are represented by grey-scale intensity value in order to avoid to use complicated unit.

3 Results and Discussion

3.1 Comparisons Between ABE and Neat Diesel

The difference of soot distribution positons and the intensity of different tested fuels, diesel, regular ABE solutions, and higher acetone contained ABE solutions (ABE(6:3:1)) is illustrated in Fig. 4. The injector produced six jets; however, only one jet was captured in order to obtain better a resolution and a higher recording speed. The injector tip and chamber wall are located in the middle, on top and bottom of each image respectively. Each row in the figure is a sequence of averaged images from

individual injections for a given fuel. It should be noted that the pixel values on the images represent the soot intensity, with crimson meaning strong soot emissions while blue means weak soot emissions.

As seen from the figure, it is deciphered that the soot producing region and the intensity of ABE is significantly lower than that of neat diesel under the tested condition, Oxy 21%@1100K. In the first row of diesel fuel, soot is apparently generated during the combustion process, with it first appearing near the injector tip, then propagating downstream and impinging on the chamber wall, finally forming an arc shape soot emission region. Overall, the development of the soot cloud is similar to that of a natural flame [29]. However, the soot produced very little at the bottom area near the wall for ABE and ABE(6:3:1), especially for ABE(6:3:1). In addition, by comparing the soot producing position between diesel and ABE, it is found that soot initially appeared closer to the chamber wall for ABE than that of diesel, and the combustion of ABE with a higher ratio of acetone almost achieves soot free combustion. This is because the viscosity is higher for diesel, thus the atomization is poor, and the mixture formed is rich. Meanwhile, because of the higher cetane number, the mixture reached the combustion condition early, thus soot produced closer to the injector. For ABE, due to the low cetane number, the preparation for the ignition is longer, so the combustion area formed at the place far from the injector tip. Due to the oxygen containing and excellent volatility of the ABE components, the mixture in the combustion region is lean, which largely suppressed the soot formation. Moreover, the spray combustion of

ABE(6:3:1) could obtain more cleaner combustion for its higher volatility due to the extra addition of acetone.

3.2 Under Varied Ambient Temperatures

Temperature is a parameter related to the soot producing and soot oxidation rate, which is able to influence the soot level during the whole spray combustion process. The soot cloud development of diesel and ABE at oxygen concentration of 16% under different ambient temperatures are compared and illustrated in Fig. 5. It can be found that, with the decrease ambient temperature, the soot level of spray combustion of diesel and ABE are both reduced. This indicates that in the range of the tested ambient temperatures, both the soot producing rate and the soot oxidation rate are decreased, but the magnitude of decrease for soot production is larger than that of soot oxidation.

Comparing the soot clouds of diesel in Fig. 5-a, and the soot clouds of ABE in Fig.5-b, it can be determined that not only the ABE produced a much smaller soot region than that of diesel, which is similar to the finding in Fig.4, but ABE also achieved soot free combustion at higher ambient temperatures. Spray combustion of diesel still forms apparent soot clouds when the ambient temperature is as low as 900K which is at least three times larger than that of ABE at the highest tested ambient temperatures, while the soot signal almost disappeared in the camera view field when the ambient temperature is reduced down to 1000K. In addition, the soot survival duration of ABE is much shorter than that of diesel, which is also beneficial to engine-out soot reduction.

In addition, the evolutions of soot clouds are different between the spray combustion of ABE and diesel. Under the higher ambient temperatures, such as 1200K and 1100K, the soot initially formed very close to the injector tip for diesel, and then propagates downstream but the soot region near the injector tip remained, thus a large soot cloud area was formed. However, with the ambient temperature reduction, the initial soot forming position of the spray combustion of diesel moved from near the injector tip to a location more downstream. When the ambient temperature was reduced down to 900K, the soot cloud of spray combustion of diesel initially formed near the chamber wall and stayed there for the entire duration of the combustion process. This phenomenon of soot forming is similar to the flame lift-off length, which increases with decreasing ambient temperature [27]. For ABE, the soot clouds stay near the chamber wall under all tested ambient temperature conditions. It is again proven that when the combustion area is far from the injector tip, the air-fuel mixing could be enhanced due to the longer journey for fuel droplet or vapor, and the sufficient oxygen will suppress soot forming and accelerate the soot oxidation.

3.3 Under Varied Ambient Oxygen Concentrations

The high temperatures and low oxygen concentrations are two essential conditions for soot forming, thus the ambient oxygen concentration is another important factor that could influence the soot level during spray combustion processes. Soot development in spray combustion of ABE and diesel under different ambient oxygen concentrations, from 21% to 11%, at high ambient temperature, 1200K, are shown in Fig. 6. The soot forming area increases with the decrease of ambient oxygen

concentrations at high ambient temperatures for both ABE and diesel. Unfortunately, the increased scale of soot region for diesel is significant larger than that of ABE. At the ambient temperature of 1200K, the soot cloud area increased largely when the ambient oxygen concentration decreased from 21% to 16%. At the same time, the intensity of the soot clouds also increased. It should be noted that there is a breach in soot clouds in the images after 4.04ms in the second row images of Fig. 6-a, under oxygen concentration of 16%. The breach should have been filled with soot signal, but counted as background due to the corresponding place on the light diffuser was contaminated by soot particles from the former cycles. The breach is bigger when testing the injection under Oxy 11%, which is why they are not shown in the figure. However, for ABE, when the ambient oxygen concentration reduced from 21% to 16% the soot cloud area increase is limited. Only when the ambient oxygen concentration further reduced to 11% soot clouds increased noticeably near the chamber wall. The magnitude of increase is still much smaller than that of diesel when the ambient oxygen concentration varied from 21% to 16%. These indicate that the advantages of ABE, such as molecule oxygen containing, high volatility, and low cetane number, could decrease its sensitivity to oxygen concentration change, and ensure clean combustion in a greater range.

3.4 Mass of Soot

According to the method shown in section 2.2, instantaneous soot mass can be obtained based on the soot density and pixel size. Hence, a more intuitive comparisons on soot emission between spray combustion of diesel and ABE are illustrated in Fig.7. the curves show that the transient soot mass inside cylinder for diesel is significantly

higher than that of ABE, especially than ABE with high fraction of acetone, which is consistent with the finding based on the images. Besides, the soot of diesel survives much longer in cylinder than that of ABE.

The soot mass of ABE spray combustion under different tested ambient temperatures and ambient oxygen concentrations are shown in Fig.8. It can be observed that there is an accumulating process in the initial process of combustion and a dissipating process in the end of the combustion for soot mass. During the former processes, the soot producing rate is much higher than the soot oxidation rate, thus the soot could accumulate rapidly and thus a soot cloud was formed. Though with the development of combustion, when the spray is finished the droplet or fuel vapor could access more oxygen. As a result, the temperature is also higher due to the main combustion, which led to a soot oxidation dominated process. As for a practical engine, the soot oxidation dominated process occurs at the expansion stroke, where the in-cylinder temperature reduces with the piston going down. Soot oxidation may be frozen when the temperature is low enough, thus there are still soot particles existing in the exhaust gas. As for constant volume chamber, the high temperature environment can remain longer, and the soot can be oxidized fully, though an amount of soot particles still adhered on the chamber wall and optical window due to their cooling effect.

From the figure, it is observed that the soot mass increases largely with decreasing ambient oxygen concentration when the ambient temperature is high, 1200K. However, it is not consistent with the variation trends of natural flame luminosity that was used to roughly represent the soot level in Ref. [29]. In the study, it was found that the natural

342 flame luminosity increased lightly with oxygen reduction for neat diesel, but decreased
343 lightly with oxygen reduction for ABE. That is different from the influence of
344 temperature on soot incandescence. The temperature of combustion area is reduced
345 when the oxygen concentration reduced, which further lowered the intensity of the
346 natural flame luminosity caused by soot particles largely. Even though the soot mass of
347 ABE increases with oxygen reduction at 1200K, but from Fig.6 it can be observed that
348 the scale of increase for diesel is much larger than that of ABE. When the ambient
349 temperature reduced down to 1100K, the soot mass of ABE almost remained at the
350 same level under varied oxygen concentration conditions. When the ambient
351 temperature is further reduced down to 1000K, the soot mass of ABE decreased with
352 the reduction in oxygen concentration, and obtained nearly free soot combustion at the
353 condition of Oxy 11%. This phenomenon is consistent with the observation based on the
354 natural flame luminosity, which is that ABE could obtain free soot combustion at the
355 condition of low temperatures and low oxygen concentrations. As a consequence, the
356 variation trend of natural flame luminosity is different to that of soot mass, but is still
357 able to predict the overall trends of soot level resulting from different fuels.

358 In addition, it also can be seen that the soot mass increased with the increase of
359 ambient temperature under all tested ambient oxygen concentrations, which is
360 consistent with the finding shown in Fig.5. However, the scale of increase under the high
361 oxygen concentrations is lower than that at the lower oxygen concentrations. This is
362 possible because the soot producing rate can be increased largely when elevating
363 temperature, but the soot oxidation rate cannot be lifted due to the insufficient oxygen.

3.5 Soot Forming Discussion

Fig. 9 illustrated the natural flame luminosity, CH* chemiluminescence, and soot distribution images of spray combustion of ABE at environments of 1200K@Oxy 21%. It should be noted that the three kinds of signals were not recorded by “tri-colors” technology, but from three injections under corresponding test systems. Inevitably, there are some deviations between signals of flame in position and intensity, which is the result of variation on in-cylinder turbulence formed by injection or heat release. CH* forming region typically was regarded to represent the rich combustion area, where the chemical reaction rate and heat release rate is high [42].

From the Fig. 9, it can be observed that natural flame luminosity initially formed closer to the injector tip than CH* and soot, and then propagates downstream and impinges on the chamber wall. The development of CH* is similar to that of natural flame, but the region is much smaller. It only matched well with the bright area in the natural flame images, which is the rich combustion area. From the images, it is observed that rich combustion usually happened in the place near the chamber wall or in the middle of the flame because the fuel there is not able to easily access oxygen there. When comparing them to the distribution of soot, it is found that soot only forms near the chamber wall, and the area is even smaller than that of the distribution of CH* signal. This phenomenon indicates that rich area in the flame could form soot, but the condition is the concentration should be high enough. Pickett et al [31], also reported if the overall equivalence ratio in the flame lift-off area is lower than around 2, soot-free combustion would be obtained.

The low soot characteristics for the spray combustion of ABE is related to its relative lean combustion. On the one hand, the viscosity of each components of ABE is lower, so the size of fuel droplet formed by atomization is reduced, which is helpful to evaporation and transportation. On the other hand, the evaporation rate itself for ABE is high due to its great volatility. In addition, as seen from Table 1, the stoichiometric air-fuel ratio of ABE (10.5) is much lower than that of diesel fuel (14.3), which means much leaner mixtures can be obtained in the same atomization condition. This explained the reason for clean combustion of ABE in one aspect. However, the soot forming also relates to the molecular structure of fuel closely. A phenomenological soot model proposed by Zhao et al [43], regarded that the main pyrolysis products in the initial air-fuel reactions include not only hydrocarbons, such as CH_4 , C_2H_2 , but also CO by considering the practical reaction processes. Harper et al [44], found that CO accounts 30% of all products in the pyrolysis process, and CO appeared earlier than C_2H_2 while temperature increased, which indicated the active energy of CO is lower than that of C_2H_2 . C_2H_2 is one of the main initial species that possibly evolves into soot particles. Therefore, the existence of a large fraction of CO in the reaction products could reduce the quantity of C_2H_2 , which would largely reduce the soot formation.

4 Conclusions

Based on a constant volume chamber system, the soot distribution of ABE and diesel spray combustion was measured through forward illumination light extinction technology. A large range of environmental conditions include ambient temperatures and oxygen concentrations corresponding to normal and low temperature modes

respectively was considered to understand the soot characteristics of ABE fully. Based on the experimental results, the soot forming mechanism of ABE was also discussed. The main conclusions are shown as follows:

- 1) Based on the experimental results of forward illumination light extinction technology, ABE, especially the one with high acetone fraction, is able to achieve much lower soot level than that of diesel.
- 2) The soot cloud region and intensity of both ABE and diesel reduced with decreasing ambient temperature, but the reduction for ABE is much larger than that for diesel, and ABE achieves soot-free combustion under higher ambient temperature conditions.
- 3) The soot cloud region and intensity of both ABE and diesel increased with decreasing ambient oxygen concentration, but the scale of increase for ABE is much lower than that for diesel, and ABE displays a low sensitivity to the oxygen concentration in the environment.
- 4) Soot mass of ABE increases with increasing ambient temperature under all tested ambient oxygen concentrations, but the scale of increase under the high oxygen concentrations is lower than that at the low oxygen concentrations because the soot production rate is increased largely when temperature is elevated but the soot oxidation rate remained due to the insufficient oxygen. However, ABE could achieve soot-free combustion at both-low (low ambient temperature and oxygen) conditions.

5) The difference between FILE soot information and natural flame luminosity are found, with soot increase with oxygen reduction at high ambient temperatures captured by FILE technology. The natural flame luminosity still can be used to predict the overall trends between fuels and test conditions.

6) Clean combustion of ABE benefits from two aspects. One is the lean air-fuel mixture in combustion region which is obtained due to the great volatility and low stoichiometric ratio. The other one is that C_2H_2 is partially replaced by CO in pyrolysis reactions that reduces the soot source materials largely.

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Tables:

Table 1 Fuel Properties

Properties	Diesel	Acetone	Butanol	Ethanol	ABE(3: 6:1)	ABE(6:3:1)
Molecular formula	C ₁₂ –C ₂₅	C ₃ H ₆ O	C ₄ H ₉ OH	C ₂ H ₅ OH		
Cetane number	40 min		25	8		
Oxygen content (wt %)		27.6	21.6	34.8	24.7	26.5
Density at 288 K (g/mL)	0.82-0.86	0.791	0.813	0.795	0.805	0.798
Auto-ignition temperature (K)	503	833	658	707		
Lower heating value (MJ/kg)	42.7	29.6	33.1	26.8	31.4	30.4
Boiling point (K)	555-561	329.2	390.8	351.5		
Stoichiometric ratio	14.3	9.54	11.21	9.02	10.5	10
Latent heat at 298 K (kJ/kg)	270	518	582	904	595	576
Flammability limits (vol. %)	0.6-5.6	2.6-12.8	1.4-11.2	4.3-19		
Saturation pressure at 311 K (kPa)	0.3	53.4	2.27	13.8		
Viscosity at 413 K (mm ² /s)	1.9-4.1	0.35	2.63	1.08		

Note: Properties of diesel are from ASTM D975; properties of acetone are from [38,39]; properties of ethanol and butanol are from [38,39,40]; the properties of ABE are calculated according to components ratios.

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577 **Table 2 Test conditions**

Fuels and ambient gas	
Tested fuels	DIESEL, ABE
Ambient density (kg/m ³)	14.8
Ambient temperature (K)	1200, 1100, 1000, 900
Ambient oxygen concentration (Vol.)	21%, 16%, 11%
Fuel injection	
Injector type	6-hole, valve-covered orifice
Orifice diameter (mm)	0.145
Injection pressure (bar)	1300
Injection duration (ms)	3.5
Injected fuel volume (mm ³)	120
High speed camera	
Resolution(pixel×pixel)	416×560
Exposure time(μs)	3
Frequency(frame/s)	9049

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581 **Figures with captions:**

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Fig. 1. Schematic of the experimental setup

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Fig. 2. Schematic of the FILE method

Fig. 3.individual and averaged soot distribution images (Diesel, Oxy 21% 1100K)

Fig. 4. Soot clouds comparison of neat diesel and ABE (Oxy 21% 1100K)

a) Diesel

b) ABE

Fig. 5. Soot clouds development of ABE and Diesel at different ambient temperatures (Oxy 16%)

a) Diesel

b) ABE

Fig. 6. Soot clouds development of ABE and Diesel at different ambient oxygen concentrations (1200K)

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Fig. 7. Soot mass comparison of neat diesel and ABE (Oxy 21% 1100K)

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Fig. 8. Soot mass development of ABE with different ambient conditions

Fig. 9. Comparison of natural flame, CH* chemiluminescence and soot distribution (Oxy 21%, 1200K)