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To FUEL*

Title:
Effects of ethyl tert-butyl ether addition to diesel fuel on characteristics of combustion and exhaust emissions of diesel engines

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Effects of Ethyl tert-Butyl Ether Addition to Diesel Fuel on Characteristics of Combustion and Exhaust Emissions of Diesel Engines

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Abstract

The effects of ethyl tert-butyl ether (ETBE) addition to diesel fuel on the characteristics of combustion and exhaust emissions of a common rail direct injection diesel engine with high rates of cooled exhaust gas recirculation (EGR) were investigated. Test fuels were prepared by blending 0, 10, 20, 30 and 40 vol% ETBE to a commercial diesel fuel. Increasing ETBE fraction in the fuel helps to suppress the smoke emission increasing with EGR, but a too high fraction of ETBE leads to misfiring at higher EGR rates. While the combustion noise and NOx emissions increase with increases in ETBE fraction at relatively low EGR rates, they can be suppressed to low levels by increasing EGR. Though there are no significant increases in THC and CO emissions due to ETBE addition to diesel fuel in a wide range of EGR rates, the ETBE blended fuel results in higher aldehyde emissions than the pure diesel fuel at relatively low EGR rates. With the 30% ETBE blended fuel, the operating load range of smokeless, ultra-low NOx (<0.5 g/kW·h), and efficient diesel combustion with high rates of cooled EGR is extended to higher loads than with the pure diesel fuel.

Keywords: Diesel Fuel; Additives; Diesel Combustion; Exhaust Emissions; Exhaust Gas Recirculation
1. Introduction

To meet the increasingly stringent regulations for exhaust gas emissions from diesel vehicles, exhaust gas aftertreatment has become a necessity in most industrialized countries. Meanwhile, from the viewpoint of maintaining the functioning of exhaust gas aftertreatment devices and fuel economy in terms of vehicle running cost, a further reduction in the engine-out emissions through advanced fuel and combustion technologies is urgently required. Conventionally, there are relatively less THC (total hydrocarbon) and CO (carbon monoxide) emissions in the exhaust gas from diesel engines than from gasoline engines, and the challenge in achieving clean diesel combustion is to simultaneously reduce NOx (nitrogen oxides) and smoke emissions.

Exhaust gas recirculation (EGR) is very effective to reduce NOx, but because of the trade-off between NOx and smoke emissions, EGR is conventionally limited to relatively low rates despite the potentials for further reduction of NOx emissions. In recent years, several concepts for premixed low temperature diesel combustion have been proposed and investigated to simultaneously reduce NOx and smoke emissions at institutions worldwide. Kitamura et al. developed a concept where NOx is reduced by a large quantity of EGR and smoke is suppressed by retarded fuel injection to elongate ignition delay as well as with a very high swirl ratio to enhance fuel-air mixing [1]. In contrast, some other researchers adopted a strategy of early fuel injection combined with very high rates of EGR to simultaneous reduce NOx and smoke emissions [2-5]. Furthermore, the effects of injection timing, injection pressure, and multiple fuel injections on the premixed low temperature diesel combustion were investigated [6-8], and study on a fundamental diesel flame in low oxygen environment was also conducted [9]. In these researches, however, the premixed low temperature combustion suffers from great increases in THC and CO emissions, and it is generally limited to low load operations. At higher loads, the increased fuel injection quantity needs more time to mix with air before ignition, while ignition delay tends to decrease due to the increased in-cylinder temperature. A further reduction in in-cylinder oxygen concentration by EGR may help to extend ignition delay, but this strategy would result in further increases in THC and CO emissions, and unacceptable deterioration in combustion efficiency. Therefore, it is extremely difficult to resolve the trade-off between NOx and
smoke emissions at higher loads while maintaining high thermal efficiency only by means of engine combustion technologies.

Fuel properties influencing engine combustion and exhaust emissions have been extensively investigated. Miyamoto and Ogawa et al. reported that an increase in oxygen content of fuels can dramatically reduce smoke, enabling more extensive utilization of EGR to control NOx emissions [10-11]. In addition, it is known that a decrease in fuel cetane number leads to elongation in ignition delay, and further the present authors found that when increasing EGR, the effect of decreased cetane number on elongating ignition delay becomes more pronounced, allowing more time for fuel-air mixing before ignition [12-13]. Moreover, Senda et al. demonstrated that addition of low boiling point component into diesel fuel promotes spray evaporation and mixture formation processes [14].

Ethyl tert-butyl ether (ETBE) can be synthesized by reacting bio-ethanol (47% v/v) and isobutene (53% v/v) with heat over a catalyst, it can be considered a “bio-fuel”, therefore ETBE helps to reduce the vehicle-out carbon dioxide (a greenhouse gas) introduced to the atmosphere. As an additive to gasoline, ETBE has been extensively examined with regard to its impact on exhaust emissions, exhaust gas aftertreatment systems, evaporative emissions, cold startability, materials used in the fueling systems and others in spark ignition engine-powered vehicles [15-17]. The fundamental characteristics regarding to ignition and combustion of both the pure ETBE and ETBE blended fuels have been studied as well [18-20]. ETBE has the properties of low auto-ignitability, low boiling point, oxygenated, and infinite solubility in diesel fuel. Therefore, ETBE, as an additive to diesel fuel, has the potentials for suppression of the smoke emissions increasing with EGR and extending smokeless and low NOx diesel combustion to higher loads by promoting fuel-air mixing as well as by its oxygenated property. Nevertheless, some concerns should be addressed when using ETBE as an additive to diesel fuel. For instance, the lowered fuel cetane number due to addition of ETBE causes a too high rate of in-cylinder pressure rise and deteriorate thermal efficiency or fuel economy. In addition, it is concerned that addition of ETBE to diesel fuel, like ethanol addition to diesel fuel [21-22], might cause some increases in unregulated toxic emissions such as carbonyl or aldehyde emissions. However, so far only few published reports with respect to ETBE as an additive to diesel
fuel are available [23], and more extensive studies regarding to the effects of ETBE addition to diesel
fuel on engine performance and exhaust emissions are needed before it can be widely used.

The objective of this study is to investigate the effects of ETBE addition to diesel fuel on the
combustion and exhaust emissions of a common rail direct injection diesel engine with high rates of
cooled EGR. In addition, the operating load range of smokeless, low-NOx, highly-efficient diesel
combustion with ETBE blended diesel fuel is discussed.

2. Experimental

2.1 Engine and operating conditions

Table 1 shows the engine specifications and operating conditions. Experiments were conducted on
a single cylinder, naturally-aspirated, four-stroke, direct injection diesel engine with a common rail
system. A disk peripherally marked by 1mm-width slits at every 20 degree crank angle was installed
on the engine output axle, and the crank angle signal was obtained by using a phototransistor (PD32:
NEC) reading the slit. The in-cylinder pressure was measured by a piezo-type pressure pick-up
(6061B: KISTLER), and along with the injector needle lift signal, it was monitored by an oscilloscope.
The top dead center was determined by the peak pressure when motoring the engine. The ignition
delay increases with EGR, and the injection timing was advanced so that the ignition timing was
maintained at top dead center (TDC). Here the ignition timing is an approximation and estimated by
the apparent change of the rate of pressure rise in the experiments. The fuel injection pressure was
set at 120 MPa. The swirl and compression ratios were 2.2 and 16, respectively. The fuel injection
quantity (mm³/stroke) was set so as to keep the same total energy input $Q_f$ (kJ/cycle) to the engine
when comparing the effects of different fuels on the engine performance and exhaust emissions. The
total energy input of 1.1 kJ/cycle corresponds to 50% load or indicated mean effective pressure
(IMEP) of 0.5 MPa under no EGR condition. The engine speed and coolant temperature were fixed
at 1320 rpm and 80°C, respectively. The EGR was realized by diverting part of the cooled exhaust
gas into the intake port with gate valves. The EGR rate is defined by

$$EGR\ rate = \frac{M_{air0} - M_{air}}{M_{air0}}$$ (1)
where $M_{\text{air}}$ and $M_{\text{air}}$ are mass flow rates of the intake fresh air without and with EGR. The flow rate of intake fresh air was obtained by using a manometer to measure the pressure difference between the inlet and outlet of an orifice on the intake surge tank. The EGR gas was cooled by a hand-made cooler with water as coolant. The temperature of the mixture of the intake fresh air and EGR gas was $30\pm 3\, ^\circ C$ under all operating conditions. When increasing EGR, there is a direct correspondence between EGR rate and intake oxygen concentration, and the intake oxygen concentration were measured with a paramagnetic-type portable oxygen tester (POT-101: SHIMAZU). The rate of heat release (ROHR) was calculated from the in-cylinder pressure data and it is an average of 45 cycles.

2.2 Property of fuel used in experiments

A commercial diesel fuel (JIS#2) for Japanese market was used in this study. Table 2 shows the properties of the diesel fuel and ETBE. Test fuels were prepared with blending ETBE to the diesel fuel by 0, 10, 20, 30 and 40% in volume. Figure 1 shows the cetane number of the test fuels. Here the cetane number was calculated based on the volumetric fractions. Menezes et al. demonstrated that the cetane number measured with ASTM D 613 shows linear correlation with the ETBE volumetric fraction in the blends [23], and an examination showed that the method based on volumetric fraction can work well as a reasonable estimation for the cetane number of ETBE blended diesel fuels. In addition, the changes of density, kinetic viscosity and distillation temperature due to the ETBE addition are expected to influence the spray and mixture formation processes.

2.3 Exhaust gas sampling and analysis

The exhaust gas was sampled about 1500 mm downstream from the engine exhaust outlet, and it was analyzed with an automotive exhaust gas analyzer (MEXA-8120: HORIBA) including NDIR (non-dispersive infrared absorption) for CO and CO$_2$, CLD (chemical luminescence detector) for NOx, and HFID (heated flame ionization detector) for THC. The smoke concentration was measured by a Bosch-type smoke meter (DSM-20AN: ZEXEL). The base value is -3.5%, thus negative value is obtained when there is no smoke or/and smoke concentration is very low. The unregulated toxic gas emissions including aldehydes, 1,3-butadiene and aromatics were measured by using a Fourier transform infrared spectroscopy (FTIR) (MEXA-4100 FT: HORIBA).
3. Results and discussion

2.1 Effect of ETBE addition to diesel fuel on combustion and exhaust emissions

Figure 2 shows the effect of large rates of cooled EGR on the ignition delay and premixing time for three ETBE fractions: 0, 20 and 40 vol%. Here, the ignition timing is defined by the crank angle with 5% of the accumulated apparent heat release. The premixing time is defined by the period from the end of fuel injection to the onset of the ignition, and it is given by subtracting the injection duration from the ignition delay. Therefore, negative values of premixing time represent ignition during fuel injection. It has been demonstrated in the previous papers that when the premixing time is longer than a critical time, smokeless combustion can be established regardless of EGR rate, fuel injection timing, and cetane number [12-13], therefore the premixing time could be a good measure for premixed low temperature diesel combustion. As shown in Fig. 2, when the EGR rate is lower than 40%, there is no significant difference in the premixing time with varying ETBE fraction. When the EGR rate is higher than 40%, higher ETBE fraction leads to a larger increase in the premixing time with EGR. As shown in Fig. 1, the fuel cetane number decreases significantly with increasing ETBE fraction, and therefore the increase in the premixing time by EGR with higher ETBE fractions can be attributed to the decrease in fuel cetane number. It should be noted that the higher ETBE fraction results in misfiring at relatively lower EGR rates.

Figure 3 shows the effect of large rates of cooled EGR on exhaust emissions for the three ETBE fractions. With the ETBE blended diesel fuel, smoke decreases over a wide range of EGR, especially with the 40% ETBE blended diesel fuel, smokeless combustion is realized at all the EGR rates tested here. Though smoke shows the same trends with increasing EGR and peaks at 12% intake oxygen concentration for both the 0% and 20% ETBE, there is a larger EGR range of smokeless operation and a more than 50% reduction at the smoke peak with the 20% ETBE fraction. NOx increases to some degree with increasing ETBE fraction from 0 to 40% without EGR, however with the intake concentration lower than 16%, NOx almost completely disappears regardless of the ETBE fraction. The THC and CO emissions show no significant difference with various ETBE fractions, and both
increase sharply with decreasing intake oxygen concentration below 14%. It should be noted that at 14% intake oxygen, while there no significant increases in HC and CO emissions, both smoke and NOx are nearly zero with 20% ETBE.

Figure 4 shows the indicated thermal efficiency $\eta_i$ and its related influencing factors including the percentage of cooling loss to coolant in the total energy input $\varphi_{w}$, the combustion efficiency $\eta_c$, and the degree of constant volume heat release $\eta_{glh}$ under the same conditions as in Fig. 3. Here the $\eta_{glh}$ is a factor proposed by List [24], which indicates the degree of reduction of thermal efficiency when heat is released far from top dead center ($\eta_{glh}$ is unity when the Otto cycle is realized). In general, with increasing EGR, the indicated thermal efficiency shows 1~2% increases at the intake oxygen concentration higher than 14% for all three fuels. In this range, the combustion efficiency is maintained at levels above 98% while the cooling loss decreases significantly with increasing EGR, resulting in improved indicated thermal efficiency. Further increases in EGR, however, result in some decreases in the indicated thermal efficiency as the combustion efficiency deteriorates severely despite the decrease in cooling loss. However, the indicated thermal efficiency is still at levels above 40% even in the range of smokeless and low NOx combustion, as a result of competition between reduction in the cooling loss and deterioration in the combustion efficiency. At EGR rates lower than 45%, the indicated thermal efficiency is lowest with 40% ETBE blended fuel. This may be attributed to a higher cooling loss because of increases in premixed combustion. This will be discussed further when ROHR is presented.

Figure 5 shows the in-cylinder maximum rate of pressure rise $dp/d\theta_{max}$ and the engine noise under the same conditions as in Fig. 3. When the EGR rate is lower than 45%, the higher ETBE fraction is associated with a higher $dp/d\theta_{max}$ and engine noise. This can be attributed to the increased premixed combustion that will be discussed in the next paragraph. However, with further increases in EGR, both $dp/d\theta_{max}$ and engine noise decrease significantly regardless of the ETBE fraction and the increase in premixing time as shown in Fig. 2. The 40% ETBE blended diesel fuel shows lower $dp/d\theta_{max}$ and engine noise around 12% intake oxygen, and this is attributed to the slower rate of heat release with the 40% ETBE blended fuel. Note that a further decrease in the intake oxygen concentration by EGR
will lead to misfiring with the 40% ETBE blended fuel.

Figure 6 shows the in-cylinder pressure and rate of heat release (ROHR) without EGR (a) and with 50% EGR (b) for the three ETBE fractions. Without EGR, the ignition delays of the pure diesel and the 20% ETBE blended fuel show no apparent differences, and with 40% ETBE it is only one degree crank angle longer. However, there is more premixed combustion and higher peak in-cylinder pressure with higher ETBE fraction. This can be attributed to the low boiling point of ETBE promoting spray evaporation and mixture formation of the ETBE blended fuels; the increase in the premixed combustion results in more NOx emissions and more cooling loss as shown in Fig. 3, and higher combustion noise in Fig. 5. With 50% EGR, the higher ETBE fraction leads to significantly longer ignition delay. The rate of heat release shows a conventional diesel combustion shape of diffusive combustion with the pure diesel fuel, indicating the presence of a very rich mixture. A rich mixture burning in an environment lacking in oxygen would result in the very high smoke emissions as shown in Fig. 3. With the 20% or 40% ETBE blended diesel fuels and the longer ignition delays, the rate of heat release show a typical premixed charge compression ignition (PCCI) combustion shape with little diffusive combustion. Therefore the smoke reduction in Fig. 3 with the ETBE blended fuels can be attributed to the low boiling point promoting fuel-air mixing, the lowered cetane number allowing more time for fuel-air mixing, as well as the oxygenated property suppressing soot formation with ETBE addition to diesel fuel.

Figure 7 shows the effect of ETBE fraction on the premixing time and ignition delay for 0%, 45, and 50% EGR. Without EGR, the premixing time or ignition delay show little change with varying the ETBE fraction. However, with 45% and 50% EGR, the higher ETBE fraction leads to significantly longer premixing time or ignition delay.

Figure 8 shows the exhaust emissions under the same conditions as in Fig. 7. Without EGR, smoke emissions are very low and decrease slightly with increasing ETBE fraction. However, NOx shows very high values that increase significantly with ETBE fraction, due to the increased premixed combustion as discussed above. Increasing the EGR rate to 45% and 50%, NOx almost disappears and shows little difference with the various ETBE fractions. Smoke increases with EGR at the lower
ETBE fractions but it decreases with increasing ETBE fraction and disappears with ETBE fraction higher than 30% for all the three EGR rates here. There is no significant difference in THC and CO emissions with the various ETBE fractions and both increase significantly with 50% EGR.

2.2 Expansion of smokeless low NOx diesel combustion with ETBE addition to diesel fuel

The results and discussion above suggest that blending more ETBE into diesel fuel helps to suppress the increases in smoke emissions while NOx emissions are reduced with large rates of cooled EGR. However, excess ETBE results in misfiring at relatively low EGR rates. As shown in Fig. 8, smokeless operation is realized with both 30% and 40% ETBE blended fuels for all three EGR rates, therefore in this section, smokeless and ultra-low NOx low temperature combustion at higher loads and the possible operating range are discussed with 30% ETBE blended diesel fuel.

Figure 9 shows the effect of 30% ETBE addition on expanding the operating load range of smokeless, low-NOx, and efficient diesel combustion with large rates of cooled EGR. Here the overall (averaged) excess air ratio $\lambda$ is defined by

$$\lambda = \frac{A/F}{(A/F)_{st}} \quad (2)$$

where $A/F$ is the actual overall air-fuel ratio and $(A/F)_{st}$ is the stoichiometric air-fuel ratio. Increases in either fuel injection quantity or EGR rate lead to decreases in the excess air ratio.

Compared with the pure diesel fuel, the 30% ETBE blended fuel achieves smokeless and ultra-low NOx operation up to a load by IMEP of 0.67 MPa using about 5% lower EGR rates, while maintaining the indicated thermal efficiency above 40%. To suppress NOx to near zero levels, the EGR rate must be higher than 40%. At IMEP of 0.72 MPa and with the 30% ETBE blended fuel, there are some increases in both smoke and NOx emissions with 30% EGR. Further increasing EGR can reduce NOx to near zero levels, but this strategy leads to drastic increases in smoke, like the smoke behavior with the pure diesel fuel at IMEP of 0.5 MPa. Increasing EGR to a rate higher than 60%, both smoke and NOx can be reduced to near zero levels with the 30% ETBE blended fuel. However, too high rates of EGR result in the excess air ratio below 1.0 at IMEP of 0.72 MPa, causing dramatic increases in THC and CO emissions as well as unacceptable deterioration in combustion efficiency and thermal efficiency. Nevertheless, the problem of too high maximum rate of pressure rise (combustion noise)
is incurred with relatively low EGR rates at higher loads when using the ETBE blended fuel. Therefore, other strategies, for example, multiple fuel injections [9] and boosting the intake pressure [25], should be adopted and further researches are needed to address these issues when using ETBE blended diesel fuels at higher load operations.

2.3 Effect of ETBE addition to diesel fuel on unregulated toxic emissions

Figure 10 shows the effect of 30% ETBE addition to diesel fuel on THC and unregulated toxic emissions including formaldehyde (HCHO), acetaldehyde (CH₃CHO), 1,3-butadiene (1,3-C₄H₆), benzene (C₆H₆), and toluene (C₇H₈). In general, all unregulated toxic emissions correlate well with THC, that is, with EGR increasing, these emissions do not change significantly at first, and then they increase greatly with intake oxygen concentration lower than 14%. The 30% ETBE addition leads to lower concentrations for all the unregulated toxic emissions except for toluene at the EGR rate higher than 50%. When the EGR rate is lower than 50%, however, both formaldehyde and acetaldehyde show significantly higher concentrations with the 30% ETBE blended fuel than the pure diesel fuel, though THC, 1,3-butadiene, and benzene shows little difference between the two fuels. This might be attributed to the oxygenated and low boiling point properties of ETBE. The 30% ETBE addition results in about 2 ppm higher toluene at all EGR rates.

Figure 11 shows THC and the unregulated toxic gas emissions from the engine with the 30% ETBE blended fuel and large rates of cooled EGR at three loads. Both formaldehyde and acetaldehyde show no significant difference due to varying the engine load, and these emissions can be reduced efficiently by an exhaust oxidation catalyst [26]. It is noteworthy that the aromatics increase sharply with the intake oxygen lower than 12% at high load (Qf = 1.4 kJ/cycle). Aromatics are difficult to remove with the oxidation catalyst under the conditions lacking in exhaust oxygen due to high EGR [26]. Therefore, when using high EGR to lower combustion noise due to ETBE addition at high loads, the problem of increased aromatics emissions must be addressed.

4. Conclusions

The effects of ETBE addition to diesel fuel on the characteristics of combustion and exhaust
emissions of a common rail direct injection diesel engine with high rates of cooled EGR have been investigated. An appropriate ETBE addition to diesel fuel helps to suppress the smoke increasing with EGR, enabling the more extensive utilization of EGR to suppress the combustion noise and NOx emissions. Therefore, smokeless, ultra-low NOx, and efficient diesel combustion can be extended to higher loads with the ETBE blended fuel than the pure diesel fuel. With further increasing the engine load, however, the trade-offs between NOx, smoke, combustion noise, thermal efficiency appear to be problematic again, and other strategies such as multiple fuel injections, intake boosting, and exhaust aftertreatment become necessary. In addition, the issue with the increased unregulated toxic emissions due to ETBE addition must be addressed, especially at higher loads and with high rates of cooled EGR.

Acknowledgement

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Reference


Caption of tables

Table 1 Engine and operating conditions

Table 2 Fuel properties
Caption of figures

Fig.1. Cetane number changed with ETBE fraction in the blended fuel

Fig.2. Effect of large rates of cooled EGR on the ignition delay and premixing time for three ETBE fractions ($Q_f = 1.1$ kJ/cycle)

Fig.3. Effect of large rates of cooled EGR on exhaust emissions for three ETBE fractions ($Q_f = 1.1$ kJ/cycle)

Fig.4. Effect of large rates of cooled EGR on indicated thermal efficiency and related factors for three ETBE fractions ($Q_f = 1.1$ kJ/cycle)

Fig.5. Effect of large rates of cooled EGR on engine noise and the maximum rate of pressure rise for three ETBE fractions ($Q_f = 1.1$ kJ/cycle)

Fig.6. In-cylinder pressure and the rate of heat release (a) without EGR and (b) with 50% EGR for three ETBE fractions ($Q_f = 1.1$ kJ/cycle)

Fig.7. Effect of ETBE fraction on the ignition delay and premixing time for three EGR rates ($Q_f = 1.1$ kJ/cycle)

Fig.8. Effect of ETBE fraction on exhaust emissions for three EGR rates ($Q_f = 1.1$ kJ/cycle)

Fig.9. Effect of 30% ETBE addition on expanding the operating load range of smokeless, low NOx, and efficient combustion with large rates of cooled EGR ($\eta > 40\%$)

Fig.10. Effect of 30% ETBE addition on THC and unregulated toxic gas emissions from the engine with large rates of cooled EGR ($Q_f = 1.1$ kJ/cycle)

Fig.11. THC and unregulated toxic gas emissions from the engine with the 30% ETBE blended fuel and large rates of cooled EGR at three loads
Table 1
Engine and operating conditions

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Table 2
Fuel properties

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*50% distillation temperature for diesel fuel
Fig. 1. Cetane number changed with ETBE fraction in the blended fuel.

Fig. 2. Effect of large rates of cooled EGR on the ignition delay and premixing time for three ETBE fractions ($Q_r = 1.1$ kJ/cycle).
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Fig. 5. Effect of large rates of cooled EGR on engine noise and the maximum rate of pressure rise for three ETBE fractions ($Q_f = 1.1 \text{ kJ/cycle}$)
Fig. 6. In-cylinder pressure and the rate of heat release (a) without EGR and (b) with 50% EGR for three ETBE fractions ($Q_f = 1.1 \text{ kJ/cycle}$)
Fig. 7. Effect of ETBE fraction on the ignition delay and premixing time for three EGR rates ($Q_f = 1.1$ kJ/cycle).
Fig. 8. Effect of ETBE fraction on exhaust emissions for three EGR rates ($Q_f = 1.1$ kJ/cycle)
Fig. 9. Effect of 30% ETBE addition on expanding the operating load range of smokeless, low-NOx, and efficient combustion with large rates of cooled EGR ($\eta > 40\%$)
Fig. 10. Effect of 30% ETBE addition on THC and unregulated toxic gas emissions from the engine with large rates of cooled EGR ($Q_f = 1.1$ kJ/cycle)
Fig. 11. THC and unregulated toxic gas emissions from the engine with the 30% ETBE blended fuel and large rates of cooled EGR at three loads.