Influence of a Novel Mixed Dialkyl Oxalate on the Combustion and Emission Characteristics of a Diesel Engine

Ao Zhou,* Wei Guo,* Hui Jin, Yangyang Li, and Yingwu Yin

ABSTRACT: Oxygen-containing alternative fuels have excellent potential to improve diesel fuel economy and reduce particulate matter (PM) emissions. In this study, a novel mixed dialkyl oxalate (mDAO) as an additive was applied to substitute conventional diesel to investigate the effects of mDAO on the combustion and emission characteristics of a high-pressure common-rail diesel engine. The research conducted suggested that the peak pressure rise rate in the main injection stage and the peak in-cylinder pressure presented the rising tendency with the increased mass fraction of mDAO at most test conditions. With the addition of mDAO, the in-cylinder temperature ($T$) and brake thermal efficiency (BTE) were higher than that of pure diesel. When the mass fraction of mDAO in the mDAO/diesel blend was 30%, the improvement of BTE was most obvious. The ignition delay was prolonged as the mass fraction of mDAO was increased due to the lower cetane number of the mDAO. In addition, adding mDAO into diesel had an effective impact on the reduction of PM emissions, while the nitrogen oxide (NO$_x$) emissions deteriorated. These results indicate that mDAO is a great potential diesel alternative fuel.

1. INTRODUCTION

The diesel engine is one of the main power sources in the transportation industry, and research on its energy conservation and emission reduction has received more attention from researchers due to the global energy crisis and the stringent emissions regulations.1,2 Thus, the application of clean and efficient alternative fuels in diesel engines is particularly urgent and important.

In recent years, many oxygen-containing alternative fuels have been proposed, mainly including alcohols, ethers, and esters. Chen et al.3 explored the influence of methanol on the particulate matter (PM) emissions of diesel engines. The results showed that the influence of different methanol substitution percent on soot and particulate number (PN) under high load mainly depended on the intake temperature, and the high intake temperature will cause an increase in PN and soot emissions. When the methanol substitution percent was more than 20%, the methanol substitution percent had a significant effect on the reduction of soot and PN. At medium and low loads, the influence of the methanol substitution percent on the PN and soot emissions mainly depended on the direct injection timing. Taghizadeh-Alisaraeid and Rezaei-Asl4 added ethanol into diesel with concentrations of 2, 4, 6, 8, 10, and 12% to study the influence of ethanol concentration on diesel engines. The results showed that compared with pure diesel, the engine torque and power could be increased by 3.8% on average when the ethanol concentration was 6%, while the vibration of the engine block was increased. When the ethanol concentration was above 8%, the ignition delay (ID) was prolonged, the in-cylinder pressure changes increased obviously, and there was a slightly increasing knock tendency.

Luo et al.5 investigated the effect of acetone–butanol–ethanol (ABE)/diesel blend on the soot emissions of a diesel engine. The results showed that butanol and ethanol in ABE inhibited the formation of soot, while acetone in ABE deteriorated soot emissions. Kuszewski6 tested the combustion characteristics of the $n$-butanol/diesel blend in a constant volume combustion bomb. He found that the ID and combustion duration of the diesel/$n$-butanol blend increased with the increase of the $n$-butanol fraction. The ID and combustion duration of the diesel/$n$-butanol blend decreased with the increase in the ambient gas temperature. The application of alcohols could improve combustion characteristics and is beneficial for the reduction of soot emissions. However, the lower cetane number and viscosity as well as the poor miscibility with diesel restrict the application of lower

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alcohols in diesel engines when there is no additional modification of the diesel engine.

As for ethers, early research focused on dimethyl ether (DME).\(^7\)\(^9\) DME is an oxygen-containing hydrocarbon with a low carbon hydrogen ratio and no C–C bond, which effectively reduces soot emissions. At the same time, the addition of DME in diesel improves brake thermal efficiency (BTE), and further reduces hydrocarbon (HC) and carbon monoxide (CO) emissions as DME concentration increases.\(^7\) However, DME is in a gaseous state at room temperature and pressure, so it is difficult to mix with diesel. Moreover, due to the high saturated vapor pressure of DME, the DME/diesel blend is prone to generate vapor lock in the engine fuel supply pipeline, which influences the fuel supply of the diesel engine and affects the dynamic performance.\(^8\) So, polyoxymethylene dimethyl ethers (PODE) with better physical and chemical properties has been applied as an alternative fuel for diesel in recent years.

The molecular formula of PODE is CH\(_3\)O(CH\(_2\)O)\(_n\)CH\(_3\), which has high oxygen content and can be mixed with diesel in any proportion. Compared with diesel and DME, PODE has a higher cetane number, and its molecular structure contains methoxy (CH\(_3\)O) that will accelerate the consumption of soot precursor by oxygen atoms to generate a large number of hydroxyl (OH), effectively reducing the emission of PM.\(^11\) PODE is a commonly used oxygenated additive in diesel, and many studies have shown that mixing PODE with diesel could effectively improve engine efficiency and significantly reduce soot and CO emissions.\(^12\)\(^15\) Pellegrini et al.\(^16\) added a 7.5% volume of PODE into diesel to study the distributions of PN size and polyaromatic hydrocarbons, and the experimental results showed that soot and PM emissions were significantly reduced. At low speed and low load conditions, the PN with particle size smaller than 30 nm increased slightly, while the PN concentration with particle size larger than 30 nm decreased at high speed. Emissions of polyaromatic hydrocarbons increased. Liu et al.\(^17\) investigated the combustion and emission characteristics of the diesel engine fueled with PODE/biodiesel, biodiesel, and pure diesel. Compared with pure diesel, the ID and combustion phasing of pure biodiesel and PODE/biodiesel blend did not change significantly. When the volume fraction of PODE was 15% in PODE/biodiesel blend, soot emissions were reduced by 50%, and CO emissions were reduced by 20% compared to biodiesel.

Ester oxygen-containing fuels are also promising alternative fuels for diesel. Among them, biodiesel and dimethyl carbonate (DMC) are superior to diesel in some fuel characteristics and can be mixed with diesel in any proportion. Therefore, the investigations of these two ester fuels as oxygen-containing alternative fuels for diesel engines have attracted much attention. The DMC has no C–C bond and high oxygen content (53.3%) compared to diesel, which contributes to the inhibition of soot.\(^18\)\(^21\) Yang et al.\(^22\) studied the effect of DMC on the emission characteristic and brake specific fuel consumption (BSFC). They found that with the addition of DMC, the combustion phasing was delayed attributed to the lower cetane number while achieving obviously reduced PM emissions. The NO\(_x\) emission and BSFC increased with the addition of DMC. Pan et al.\(^23\) also found that the DMC had a positive effect on the reduction of soot emissions, while had an opposite effect on NO\(_x\) emissions and BTE. Compared to DMC, biodiesel is more suitable as an alternative for diesel due to its higher low heat value, kinematic viscosity, and flash point as well as a wide range of production sources.\(^24\)\(^26\) Esakkii et al.\(^27\) pointed out that the application of biodiesel in diesel engines combined with exhaust gas recirculation (EGR) could reduce PM emissions and simultaneously reduce NO\(_x\) emissions. Mourad et al.\(^28\) found that the NO\(_x\) emissions increased by using biodiesel compared with diesel. However, the use of preheated method combined with EGR could reduce NO\(_x\) emissions and improve the thermal efficiency of the engine. Rajak et al.\(^29\) studied the emission characteristics of a diesel engine fueled with microalgae biodiesel. The results showed that the PM and NO\(_x\) emissions decreased with the adoption of microalgae biodiesel compared to pure diesel.

Through a review of the abovementioned literature, it is clear that the use of oxygenated fuels has a significant effect on reducing the main emissions of diesel engines. In this study, a novel alternative fuel (mDAO), a kind of oxalate esters, was added to diesel to study the influence of mDAO on the combustion and emission characteristics of a diesel engine. The mDAO is synthesized from dimethyl oxalate and higher alcohols through solvent-free partial transesterification, and the main raw materials of dimethyl oxalate are CO-rich industrial waste gases and syngas.\(^30\) So, the cost of production of mDAO is cheap, and the mDAO is environmentally friendly. In addition, the oxygen content of mDAO is 40%. The exploitation and utilization of mDAO in diesel engines will effectively alleviate the oil crisis. At the same time, due to its high oxygen content, it can reduce PM emissions, allowing diesel engines to adapt to stricter emission regulations. The purpose of this study is to verify the potential of mDAO as a diesel oxygen-containing additive in reducing emissions and investigate its effect on the combustion characteristics of diesel engines, especially on BTE. Until now, there is no published literature about the application of mDAO in compression ignition engines. The results of this study could offer guidelines regarding the commercial application of mDAO as a diesel additive.

### 2. EXPERIMENTAL APPARATUS AND METHODOLOGY

#### 2.1. Experimental Setup

The experimental tests were conducted on a four-cylinder, high-pressure common-rail compression ignition engine. The specifications of the test engine are listed in detail in Table 1. The schematic diagram of the engine test system is shown in Figure 1. To achieve the data of cylinder pressure, a pressure transducer (Kistler, 6052A) was applied and mounted in the head of cylinder #1, and the cylinder pressure signal was processed by a charge amplifier (Kistler, 5019B) before being input to the combustion analyzer (Kibox 2893A, Kistler) for record and analysis. In addition, a current clamp (Tektronix, A622) was used to measure the in-cylinder direct injection signal of injector #1. In this study, the consumption of the tested fuels was measured by a transient

| Table 1. Engine Specifications | values |
|--------------------------------|--------|
| bore (mm)                       | 105    |
| stroke (mm)                     | 130    |
| link length (mm)                | 210    |
| compression ratio               | 18:0:1 |
| displacement (L)                | 4.5    |
| rated power (kW)@speed (rpm)   | 113@2300 |
| rated torque (Nm)@speed (rpm)   | 520@1600–1800 |
| cooling type                    | water  |
fuel consumption meter (Toceil, Toceil-CMFG010). The engine speed and load were controlled through an eddy current dynamometer.

Besides, an AVL DiGas 4000 gas analyzer was used to measure \( \text{NO}_x \) concentration and a particle counter (Longshun, CLJ-E) was applied to measure the PN in the exhaust gas. The specifications of all test equipment are listed in Table 2.

The uncertainties of the measured and calculated parameters negatively affect the accuracies of the obtained results. Therefore, the uncertainties of all the experimental data are calculated for verifying that the experiments are reliable and have high precision. In this study, the calculation method of the uncertainty is based on the study by Zhang et al., and the uncertainties of experimental parameters are presented in Table 3.

2.2. Test Fuels. In this study, two kinds of fuels, pure diesel and mDAO/diesel blend fuels, were used. For mDAO/diesel blend fuels, mDAO was blended into diesel with 10, 20, and 30% mass fraction ratio, which was defined as MAO10, MAO20, and MAO30, respectively. The properties of the four tested fuels are listed in Table 4.

2.3. Experimental Conditions. In this study, twelve engine operating conditions were selected for each tested fuel, as seen in Table 5. The pilot injection strategy was applied, and the pilot injection period was not changed under each test condition. While the pilot injection timing and main injection timing were

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**Figure 1.** Schematic diagram of the engine test system.

**Table 2. Specifications of the Test Instrument**

| measured parameter | instrument       | measuring range     | accuracy       |
|--------------------|------------------|---------------------|----------------|
| torque/speed       | FST3 CW160B      | 0–600 Nm/0–10,000 rpm | 0.22%          |
| diesel injection signal | Tektronix A622 | 0.5–100 A | ±0.01 ms |
| diesel consumption  | Toceil CMFG010 | 0–60 kg/h | ≤0.12% |
| in-cylinder pressure | Kistler 6052A | 0–25 MPa | 0.5% |
| crank angle        | Kistler 6291     | 0–720 °CA | 0.1 °CA |
| \( \text{NO}_x \)  | AVL DiGas 4000   | 0–5000 ppm          | ±1 ppm |
| particle number    | Longshun CLJ-E   | <35,000 #/L          |                |

**Table 3. Uncertainties of Parameters**

| parameters | uncertainty (%) |
|------------|-----------------|
| speed      | 0.3             |
| pressure   | 0.7             |
| temperature| 1.8             |
| fuel consumption | 3.0       |
| \( \text{NO}_x \) | 2.1         |
| BTE        | 3.7             |

**Table 4. Diesel and mDAO/Diesel Blend Properties**

| properties | diesel | MAO10 | MAO20 | MAO30 |
|------------|--------|-------|-------|-------|
| kinematic viscosity (mm² s⁻¹) | 6.202 | 5.732 | 5.178 | 4.743 |
| lower heating value (MJ/kg)    | 42.861| 40.422| 38.576| 35.918|
| cetane number                  | 55.2  | 53.4  | 50.9  | 49.4  |
| flash point (°C)               | 78.5  | 66.5  | 63.0  | 62.0  |
| oxygen content (%)             | 0     | 3.9   | 7.8   | 11.7  |

**Table 5. Test Conditions**

| engine speed (rpm) | engine torque (Nm) | main injection timing (°CA BTDC) | pilot injection timing (°CA BTDC) |
|--------------------|--------------------|----------------------------------|----------------------------------|
| 1200               | 50                 | 18.0                             | 10.0                            |
| 1200               | 100                | 18.1                             | 10.1                            |
| 1200               | 150                | 18.5                             | 10.5                            |
| 1200               | 200                | 19.0                             | 11.0                            |
| 1200               | 250                | 19.6                             | 11.6                            |
| 2000               | 50                 | 19.5                             | 10.0                            |
| 2000               | 100                | 19.6                             | 10.1                            |
| 2000               | 150                | 20.2                             | 10.7                            |
| 2000               | 200                | 21.8                             | 12.3                            |
| 2000               | 250                | 22.5                             | 13.0                            |
| 2000               | 300                | 22.7                             | 13.2                            |
determined due to the best engine power performance when the engine was fueled with pure diesel, the pilot injection timing, main injection timing, and injection pressure were not adjusted under corresponding operating conditions when the engine was fueled with the other three fuels.

3. RESULTS AND DISCUSSION

3.1. Combustion Characteristics. Figure 2 shows the effects of mDAO on the in-cylinder pressure and peak pressure rise rate (PRR) curves at 1200 rpm and under different torques conditions. As shown in Figure 2, at low and medium engine
loads, the variation trend of the in-cylinder pressure of the mDAO/diesel blend is not obvious, while the peak in-cylinder pressure of the mDAO/diesel blend is higher than that of diesel. At the same time, as can be seen from Figure 2, the PRR curve presents a tendency to move toward the top dead center (TDC) during the first combustion stage, while the PRR curve of diesel is behind that of the mDAO/diesel blend in the second combustion stage, except the highest load condition. During the first combustion stage, with the addition of mDAO and the increased mass fraction of mDAO, the cetane number of the fuel
declines, which results in the delay of ignition. For the second combustion stage, the delayed combustion phase of the mDAO/diesel blend increases the impact of released heat from the pilot injection fuel on the main combustion process, leading to the early ignition of the mDAO/diesel blend.\textsuperscript{2} In addition, because mDAO is an oxygenated fuel, it provides additional oxygen for the combustion of the in-cylinder fuel–air mixture, which results in the improved combustion process and the increased heat released from the second combustion stage, finally resulting in the peak in-cylinder pressure of the mDAO/diesel blend being higher than that of pure diesel. However, at the highest load condition, the $T$ is enough high for the ignition of the fuel–air mixture and weakens the impact of the released heat from the pilot injection fuel on the main combustion process. Therefore, the cetane number of the fuel plays a dominant role in the combustion phase of the main combustion stage. Furthermore, with the increase in engine load, the diesel consumption is equal to or even higher than that of the mDAO/diesel blend, and the lower heating value of diesel is higher. Therefore, the peak in-cylinder pressure and PRR are higher when the engine is fueled with pure diesel.

Figure 3 shows the effects of mDAO on the in-cylinder pressure and PRR curves at 2000 rpm and under different torques conditions. Compared to the test conditions at 1200 rpm, it could be found that the variation trend of peak in-cylinder pressure and PRR curves are similar. As mDAO is an oxygenated fuel, the combustion of the mDAO/diesel blend is improved compared with that of pure diesel. However, when the engine load increases, the consumption of the mDAO/diesel blend becomes lower than that of pure diesel. All these factors contribute to the decrease of difference between peak in-cylinder pressure of diesel and the mDAO/diesel blend. As seen in Figure 3, it can also be found that the PRR curves of the mDAO/diesel blend are behind that of pure diesel, which is due to the lower cetane number of the mDAO/diesel blend. Additionally, there is an obvious difference in PRR curves compared with the conditions at 1200 rpm. At 2000 rpm, the peak PRR of all tested fuels during the second combustion stage is visibly lower than that of the first combustion stage, while the
peak PRR of the mDAO/diesel blend is higher than that of pure diesel at low and medium loads. This is because, with the increase of engine speed, more heat is released from the main injection fuel during the expansion stroke, leading to less heat released before TDC (BTDC) and significantly lower peak PRR. At low and medium loads, the addition of mDAO decreases the viscosity of the fuel blend, and the atomization quality of the mDAO/diesel blend is improved. Also, the mDAO increases the oxygen content. All of these accelerate the combustion process, which increases the heat quantity released from the main injection fuel in the compression stroke, resulting in a higher peak PRR of the mDAO/diesel blend during the main combustion stage than that of pure diesel.

Figure 4 depicts the effects of mDAO on the peak $T$ under different engine torques at 1200 and 2000 rpm. With the addition of mDAO, the peak $T$ of the mDAO/diesel blend is almost higher than that of pure diesel, especially at low and medium loads. On the one hand, the lower viscosity of mDAO is beneficial for the formation of the uniform fuel–air mixture, improving the whole combustion process, especially at low loads. On the other hand, adding mDAO can increase the oxygen content of mixed fuel and reduce the probability of diesel generating soot due to local high temperature and hypoxia, facilitating more complete combustion of the mDAO/diesel blend. The peak $T$ of the two kinds of fuel increases gradually with the increase of engine torque at high- and low-speed conditions, which is related to the increased mass of fuel. Furthermore, it is noted that the peak $T$ of MAO10 is almost higher than that of the other three test fuels at low and medium loads, while the peak $T$ of MAO30 is almost the lowest. This is the result of the combined influences of the direct injection quantity, ignition ability, atomization quality, and the oxygen content of the fuel.

Figure 5 illustrates the ID of diesel and three mixed fuels under different test conditions. In this study, the crank angle corresponding to 10% of accumulated heat release is defined as CA10, and the ID is defined as the interval between the crank angle of direct injection timing and CA10. It can be seen from
the figure that the ID is significantly prolonged with the addition of mDAO under the same test condition, and the ID increases with the increased mass fraction of mDAO, which is caused by the low cetane number of mDAO. This result was affirmed by Pan et al.\textsuperscript{32} using DMC, which had a low cetane number compared to diesel. For the same fuel, the ID is shortened with the increase of engine load. The main reason is that the low $T$ during the compression stroke is not conducive to the ignition of direct injection fuel at a low load, resulting in a longer ID. When the engine load rises, the ambient temperature surrounding the fuel spray is obviously increased, which promotes the evaporation of the fuel spray. Therefore, the ID is shortened.

Figure 6 shows the variation of BTE under different engine torques for MAO10, MAO20, MAO30, and pure diesel at 1200 and 2000 rpm. Data in Figure 6 suggest that the BTE of the mDAO/diesel blend is higher than that of diesel. Because mDAO is oxygenated fuel, which reduces the local hypoxic areas, it promotes the complete combustion of the mDAO/diesel blend. Meanwhile, the viscosity of mDAO is lower than diesel, so the atomization of the mDAO/diesel blend is improved compared to that of pure diesel. Thus, the addition of mDAO can improve the BTE of diesel engines. However, at low loads, the $T$ is low. Compared to diesel, the cetane number of mDAO is lower, and the latent heat of vaporization of mDAO is higher. All of these lead to the reduction of BTE with the addition of mDAO at low loads. In addition, when the mass fraction of mDAO is 30%, the BTE is highest at medium and high loads.

### 3.2. Emission Characteristics

Figure 7 shows the variation curve of $NO_x$ emissions with load at different engine speeds. In general, the $NO_x$ emissions of mDAO/diesel blends are higher than that of pure diesel under all test conditions. The rising tendency of $NO_x$ emissions is in line with previous studies using other oxygenated fuels, such as DMC.\textsuperscript{22} First, with the addition of mDAO, the viscosity of mixed fuel decreases, and the atomization quality is improved. Second, with the addition of mDAO, the ID is prolonged, which results in the formation of a more uniform fuel–air mixture. In addition, the oxygen content of the mDAO/diesel blend increases, which accelerates the combustion process. Therefore, the average $T$ of the mDAO/diesel blend is higher than that of pure diesel, leading to an increase in $NO_x$ emissions. However, when the mass fraction of mDAO is 30% with the engine torque of 300 Nm, the $NO_x$ emissions of MAO30 are not the highest. As is known, the formation of the $NO_x$ emissions is mainly affected by $T$, oxygen concentration, and reaction residence time.\textsuperscript{33} Although the $T$ of MAO30 is highest at high loads, the improved in-cylinder thermal environment shorts combustion durations, which results in shorter reaction residence time. All these factors lead to the reduction of $NO_x$ emissions at high loads for the mDAO/diesel blend.

Figure 8 displays $PN$ concentration with the particle size over 0.3 $\mu$m under different test conditions. Diesel engine load is regulated by changing fuel injection quality, and PM is mainly generated in high temperatures and oxygen-deficient regions.\textsuperscript{11} With the increase of engine load, the direct injection fuel quantity increases, and the temperature in the cylinder increases obviously with the increase of load. Subsequently, more soot core is generated through thermal cracking of the fuel in the center of the fuel spray and other areas where the concentration of the fuel–air mixture is too high, resulting in an increase in PM emissions. Compared to pure diesel, as the mass fraction of mDAO is increased, the PN concentration presents a declining tendency. First, with the increment in the mass fraction of mDAO, the oxygen content of the mDAO/diesel blend increases, which inhibits the formation of the soot core and accelerates complete combustion. Second, with the increased mass fraction of mDAO, the viscosity of the mDAO/diesel blend decreases. Therefore, the uniformity of the fuel–air mixture is improved, resulting in the reduction of the PN concentration.\textsuperscript{11}

### 4. CONCLUSIONS

In this study, a novel oxygenated fuel (mDAO) was added to diesel to investigate its effect on the combustion and emission characteristics of a diesel engine. The results obtained from this study could offer guidelines regarding the commercial application of mDAO as a diesel additive. The main conclusions are as follows:

1. With the addition of mDAO, the peak in-cylinder pressure and $T$ are almost higher than that of pure diesel at all test conditions.
(2) The ID is gradually prolonged with the increased mass fraction of mDAO. By adding mDAO, the BTE of the diesel engine is improved. When the mass fraction of mDAO is 30%, the BTE is highest, except for the lowest load conditions.

(3) At medium and high loads, the NO\textsubscript{x} emissions of mDAO/diesel blends are higher than that of pure diesel. However, the PM emissions are reduced with the addition of mDAO under all test conditions.

(4) In terms of combustion and emission characteristics, mDAO is a great potential diesel alternative fuel.

## AUTHOR INFORMATION

**Corresponding Authors**

Ao Zhou — School of Mechanical Engineering, Lanzhou Jiaotong University, Lanzhou 730070, China; orcid.org/0000-0001-9468-7357; Phone: +86-931-4955702; Email: cn_zhouao@163.com

Wei Guo — CAS Key Laboratory of Design and Assembly of Functional Nanostructures, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou 350002, China; Phone: +86-592-2186198; Email: xmguowei@fjirsm.ac.cn

**Authors**

Hui Jin — School of Automotive Engineering, Lanzhou Institute of Technology, Lanzhou 730050, China

Yangyang Li — Shaanxi Key Laboratory of Development and Application of New Transportation Energy, Chang’an University, Xi’an 710064, China

Yingwu Yin — School of Materials Science and Engineering, University of Jinan, Jinan 250022, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.2c04477

**Notes**

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## NOMENCLATURE

- ABE, acetone—butanol—ethanol
- BSFC, brake specific fuel consumption
- BTE, brake thermal efficiency
- CO, carbon monoxide
- DMC, dimethyl carbonate
- DME, dimethyl ether
- EGR, exhaust gas recirculation
- HC, hydrocarbon
- ID, ignition delay
- mDAO, mixed dialkyl oxalate
- NO\textsubscript{x}, nitrogen oxide
- OH, hydroxyl
- PM, particulate matter
- PN, particulate number
- PODE, polyoxymethylene dimethyl ethers
- PRR, pressure rise rate
- T, in-cylinder temperature
- TDC, top dead center

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