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## IMPACT OF FORMALDEHYDE ADDITION ON AUTO-IGNITION IN INTERNAL-COMBUSTION ENGINES

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By employing a direct-injection diesel engine equipped with a common-rail type of injection system, by adding formaldehyde (CH<sub>2</sub>O) to the intake air, and by changing the fuel-injection timing, the compression ratio and the intake-air temperature, a mechanism for CH<sub>2</sub>O as a fuel additive to affect auto-ignition was discussed. Unlike an HCCI type of engine, the diesel engine can expose an air-fuel mixture only to a limited range of the in-cylinder temperature before the ignition, and can separate low- and high-temperature parts of the mechanism. When low-temperature oxidation starts at a temperature above 900 K, there are cases that the CH<sub>2</sub>O advances the ignition timing. Below 900 K, to the contrary, it always retards the timing. It is because, above 900 K, a part of the CH<sub>2</sub>O changes into CO together with H<sub>2</sub>O<sub>2</sub> as an ignition promoter. Below 900 K, on the other hand, the CH<sub>2</sub>O itself acts as an OH radical scavenger against cool-flame reaction, from the beginning of low-temperature oxidation. Then, the engine was modified for its extraordinary function as a gasoline-knocking generator, in order that an effect of CH<sub>2</sub>O on knocking could be discussed. The CH<sub>2</sub>O retards the onset of auto-ignition of an end gas. Judging from a large degree of the retardation, the ignition is probably triggered below 900 K.

**Key Words:** Internal Combustion Engine, Ignition, Combustion, Auto-Ignition, Spray Combustion, Knocking, Ignition Control, Additive, Formaldehyde, Flame Light Emissions

### 1.Introduction

There are auto-ignition dominated subjects of combustion in internal combustion engines, that is, those of combustion control for homogeneous-charge compression-ignition (HCCI) types of engines, knocking control for gasoline engines, and new combustion modes for diesel engines to reduce both NO<sub>x</sub> and smoke emissions. Especially of HCCI engines, a lot of researchers

have been making efforts to resolve one of its difficult problems, how to control the ignition timing and stabilize the sensitive timing, as they recognized its potential advantages of a high thermal efficiency and very-low NO<sub>x</sub> and zero smoke emission levels. Effects of directly-controllable factors such as fuel properties and engine-operating conditions of the intake-air temperature, the compression ratio, the air-fuel ratio and the EGR ratio on the ignition

have been investigated. As another approach to HCCI timing control, ideas of making use of a fuel additive, for example, hydrogen peroxide(1) or ozone(2) as an ignition promoter, and formaldehyde(3) – (5) or methanol(2) as an ignition retardant, have been proposed. By Aceves et al. with the aid of calculation involving one-dimensional hydrodynamics transport and detailed chemical kinetics, hundreds of potential additives were ranked according to their capabilities for advancing the ignition timing(6).

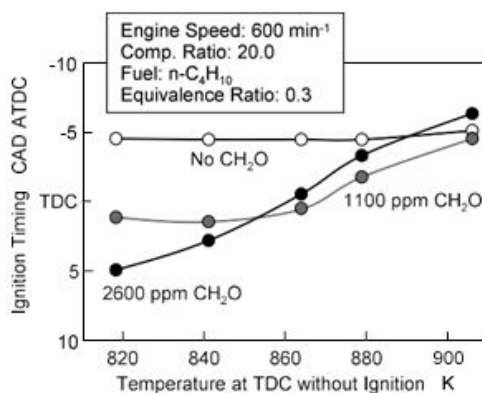


Fig. 1 Effect of CH<sub>2</sub>O addition on ignition timing of air n-C<sub>4</sub>H<sub>10</sub> mixture<sup>(4)</sup>

## 2. Experimental Procedure

### 2.1 Test engine and its operating conditions

A four-valve single-cylinder naturally-aspirated direct-center-injection diesel engine equipped with a common-rail type of injection system, was supplied for this study. A pressure transducer (AVL GU12S-10) was mounted on the cylinder head, in place of the glow plug. The intake-air temperature was measured 70 mm upstream of an intake valve. A part of the exhaust gas was by-passed into an exhaust-gas analyzer (Horiba MEXA-8220D). Table 1 lists specifications of the engine together with its

operating conditions. In a conventional use of the engine fueled with gas oil (cetane index (JISK2280): 52), the compression ratio was set at 18.0 or

12.3. The fuel-injection timing was changed between 30 and 5 crank-angle degrees (CAD) ATDC. The injection timing of 30 CAD ATDC is close to the limit of the advancement, for fuel sprays out of the multi-hole nozzle to be captured within the piston cavity. But also, conveniently, it is almost coincident with a timing before which any cool flame could not appear at the low compression ratio, even in case of HCCI. There was also a case at the low compression ratio, that the intake-air temperature was increased by 50 K using a heater installed upstream of the intake manifold, in order that the in-cylinder temperature at TDC only with the air could be almost the same as that at the high compression ratio without any heating. In these ways, a temperature to which an air-fuel mixture was exposed before the auto-ignition, was changed between 660 and 930 K at a rough estimate with the intake-air temperature without any consideration of a residual gas and a heat loss. In another case at the low compression ratio, O<sub>2</sub> was added to the intake air, and its level in the charge was increased up to 26.0%, so that the difference could be discussed, when the ignition delay was shortened by the high temperature or by the high O<sub>2</sub> level.

Table 1 Engine specifications and operating conditions

	CI Diesel Combustion	SI Gasoline Combustion
Bore / Stroke / Displacement	91.1 mm / 95.0 mm / 619 cm <sup>3</sup>	
Engine Speed	1200 min <sup>-1</sup>	
Comp. Ratio (Geometrically)	12.3 (11.4)	18.0 (16.7)
Combustion Chamber	Reentrant Type of Cavity (Min. Dia. / Max. Dia. / Depth: 46.0 / 54.9 / 23.2 mm)	Reentrant Type of Cavity (Min. Dia. / Max. Dia. / Depth: 44.2 / 49.3 / 18.2 mm)
In-Cylinder Flow	Swirl-Control Valve Closed (Estimated Swirl Ratio of 3.7 at Differential Press. of 0.004 MPa through Steady-Flow Rig)	
Fuel	Gas Oil (Cetane Index (JISK2280): 52)	Regular Gasoline (RON: 90.3)
Fuel Preparation	φ 0.16-5-Hole Nozzle, Common-Rail Press.: 50 MPa	Intake-Port Injector
Location of	Nozzle: on Cylinder Axis	Plug: 18 mm off Cylinder Axis
Timing of	Injection: -30, -20, -10 and -5 CAD ATDC	Ignition: -10 CAD ATDC
Air Charge	NA, WOT, No EGR	65% Charge, No EGR
Charge Temperature in Intake Port	305 K, 355 K (Heating)	305 K, 312 K (35% Charge)
Coolant Temperature	358 K	
O <sub>2</sub> Level in Charge	Atmospheric, Rich (26 %)	Atmospheric
CH <sub>2</sub> O Level in Charge	1900 +/- 100 ppm	1900 +/- 100 ppm, 5000 ppm (35% Charge)
Air or O <sub>2</sub> Excess Ratio Excluding CH <sub>2</sub> O	3.4 (Heating), 4.2 (26% O <sub>2</sub> )	3.4, 1.2 (35% Charge)
Temperature at Injection Timing	610, 700, 770 and 800 K, 720, 810, 900 and 930 K (Heating)	660, 770, 880 and 930 K

## 2. 2 Formaldehyde feed

CH<sub>2</sub>O-feeding conditions were set equally to those in the previous studies. In the studies, a part of in-cylinder gas had been directly sampled, and the level of CH<sub>2</sub>O in it had been measured using a gas chromatograph equipped with a flame ionization detector (GL Science GC-390TTDF)(3), (4). The previous data could be used for calibration on the level of CH<sub>2</sub>O in this study.

The amount of CH<sub>2</sub>O was monitored also by means of its value derived from the difference in the total amount of C atoms in exhaust emissions with or without the CH<sub>2</sub>O addition. Its validity is dependent on a previous result that, even in an extremely-lean HCCI engine, almost all of added CH<sub>2</sub>O is oxidized into CO and CO<sub>2</sub> during the combustion, which are detectable using the exhaust-gas analyzer, as shown in Fig. 2(4). Actually, however, in many cases at the low compression ratio in the conventional use of the engine, the indicated total level of HC emissions with the CH<sub>2</sub>O was higher than that without any addition.

The difference was less than 200 ppm, although the value is unreliable.

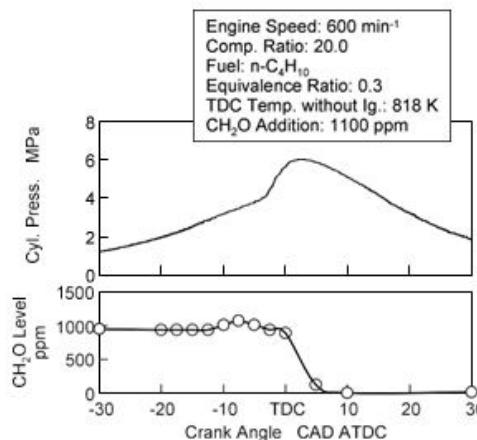


Fig. 2 History of in-cylinder CH<sub>2</sub>O level in case of HCCI engine fueled with n-C<sub>4</sub>H<sub>10</sub>(4)

a momentary engine operation at the high O<sub>2</sub> level. After all, there was no big difference in the value based on the C counting or the calibration with the previous data.

In some engine-operating conditions, the indicated total level of HC emissions with the CH<sub>2</sub>O addition was higher than that without any addition, although the value of the difference is unreliable. The CO level with the CH<sub>2</sub>O was always higher than that without any addition. A part of the CH<sub>2</sub>O, distributed in local areas off from combustion, seems to be emitted as being something ex- cept for CO<sub>2</sub> and H<sub>2</sub>O.

## 3. Results and Discussion

### 3. 1 Effect on formaldehyde addition on auto- ignition in diesel engine

Figures 3 to 5 show histories of the in-cylinder pres- sure and the effective heat release rate (EHRR) with or without the CH<sub>2</sub>O addition, at the different compression ratios and charge temperatures,

acquired every 0.125 CAD over continuous 30 cycles and cycle-averaged. Figure 6 shows ignition delays from the fuel-injection timings. Here, the ignition timing was defined as the timing when the gradient of an EHRR history first reaches 0.02 MPa/CAD<sup>2</sup> (12.4 J/CAD<sup>2</sup>) in an empirical way. Cool-flame reaction or faint heat release before the ignition appears at the same timing with or without the CH<sub>2</sub>O, as drawn with a single dotted line. At the low compression ratio, the CH<sub>2</sub>O weakens the pre-ignition heat release, and retards the ignition timing with any injection timings, compared to that without any addition. This is remarkable with the earliest injection timing when a two-stage type of heat release characterized by cool-flame reaction, appears.

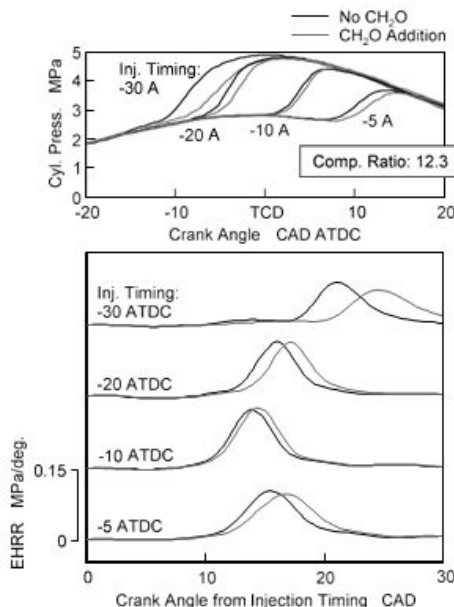


Fig. 3 Histories of in-cylinder pressure and effective heat-release rate in case of low compression ratio

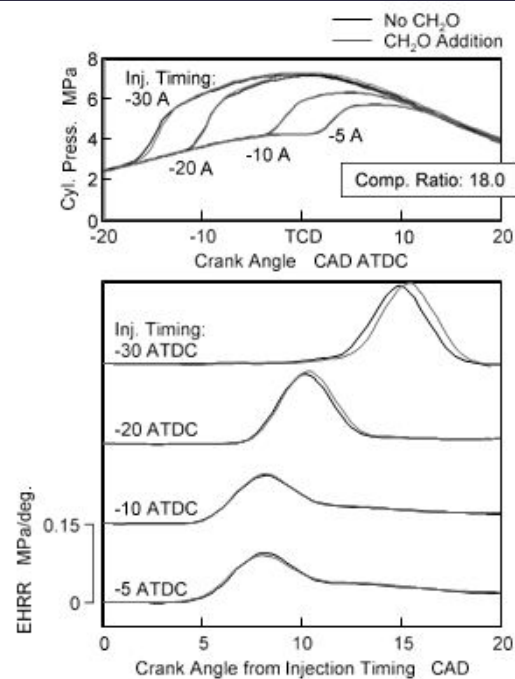


Fig. 4 Histories of in-cylinder pressure and effective heat-release rate in case of high compression ratio

### 3. 2 When ignition delay is decreased

For an extended understanding of the opposite effects of the CH<sub>2</sub>O addition, contrastive cases to the above ones were discussed. One that the ignition delay was shortened by the O<sub>2</sub> addition to the intake air, even at the low compression ratio without any assistant rise in the charge temperature, to the extent of the delay shortened by the charge heating. The other that the delay was extended by the intake-air throttling, even at the high compression ratio, to the extent of that at the low compression ratio.

Figure 7 shows ignition delays as well as histories of the in-cylinder pressure and the EHRR, at the high O<sub>2</sub> level in the charge.

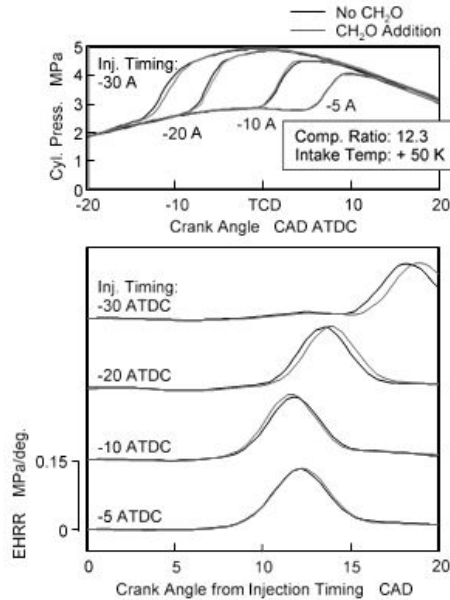


Fig. 5 Histories of in-cylinder pressure and effective heat-release rate in case of low compression ratio and heating

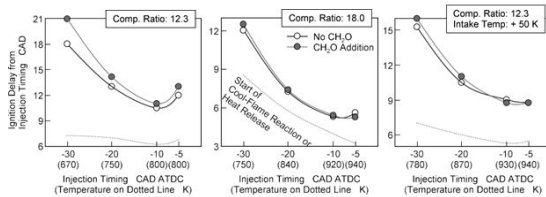
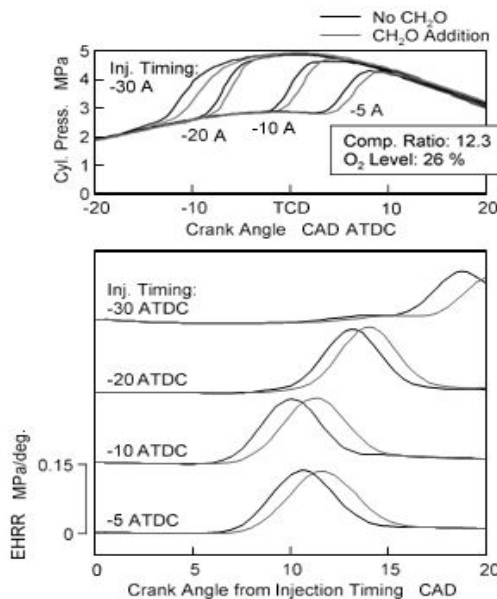


Fig. 6 Effect of CH<sub>2</sub>O on ignition delay



(a) Histories of in-cylinder pressure and effective heat-release rate

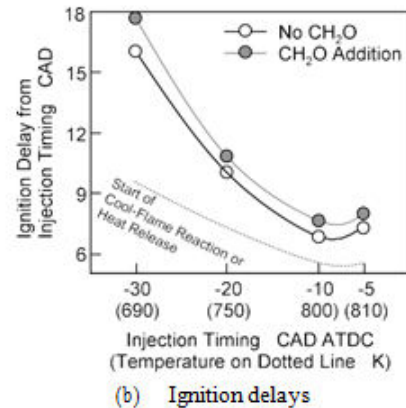


Fig. 7 Effect of CH<sub>2</sub>O on ignition delay in case of low compression ratio and high O<sub>2</sub> level

## 4. Conclusions

For close discussion about a mechanism for CH<sub>2</sub>O as a fuel additive to interact with auto-ignition of an air-fuel mixture, a direct-injection diesel engine equipped with a common-rail type of injection system was employed. It is because, unlike an HCCI engine, it can expose a mixture only to a limited range of the in-cylinder temperature before the ignition, and therefore, can separate low-temperature and high-temperature parts of interaction of CH<sub>2</sub>O, that is, parts interacting with cool-flame reaction and blue-flame reaction. Vapor of CH<sub>2</sub>O was added to the intake air. The ignition was induced at different fuel-injection timings, compression ratios and intake-air temperatures, that is, at different in-cylinder temperatures. Light emission spectra at the ignition timing were detected. When low-temperature oxidation starts at a temperature above 900 K, there are cases that the CH<sub>2</sub>O addition advances the ignition timing. Below 900 K, to the contrary, it always retards the timing. In case of the remarkable retardation, the

CH<sub>2</sub>O completely eliminates the OH radical emission. After all, the experimental results of the ignition timing are consistent with those using an HCCI engine in Fig. 1. This means that in case of the advancement of the HCCI timing, the interaction of CH<sub>2</sub>O with cool-flame reaction is negligible for the timing.

The opposite effects of the CH<sub>2</sub>O on the ignition delay were most simply modeled. Above 900 K, a part of the CH<sub>2</sub>O is changed into CO and H<sub>2</sub>O<sub>2</sub>, and the latter acts as an OH radical generator in the final stage of low-temperature oxidation. Below 900 K, on the other hand, the CH<sub>2</sub>O itself acts as an OH radical scavenger against cool-flame reaction, from the beginning of low-temperature oxidation.

Then, the engine was modified for its extraordinary function as a gasoline-knocking generator, in order that an effect of CH<sub>2</sub>O on knocking could be discussed. The CH<sub>2</sub>O retards the onset of auto-ignition of an end gas. Judging from a large degree of the retardation, the ignition is probably triggered below 900 K.

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