

Fuel-Dithering Optimization of Efficiency of TWC on Natural Gas IC Engine

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ABSTRACT

Steady-state, transient and dithering characteristics of emission conversion efficiencies of three-way catalysts on natural gas IC engine were investigated experimentally on a single-cylinder CFR engine test bench. Steady-state runs were conducted as references for specific engine emission levels and corresponding catalyst capacities. The steady-state data showed that conversion of HC will be the major problem since conversion of HC was effective only for a very narrow range of exhaust mixture. Unsteady exploration runs with both lean-to-rich and rich-to-lean transitions were conducted. These results were interpreted with a time scale analysis, according to which a qualitative oxygen storage model was proposed featuring the difference between oxygen absorption and desorption rates on the palladium catalysts. Additionally this oxygen storage model also revealed information about parameter selection for dithering to achieve optimal emission operation where all three, HC (mostly CH_4), CO and NO_x are simultaneously reduced.

Dithering with different midpoint Lambda values, frequencies and amplitudes were further investigated. The results showed that the optimal dithering Lambda midpoint resided in slightly rich engine charge. An optimal intermediate frequency was found corresponding to the characteristic time of the emission-favorable Lambda zone as suggested by the oxygen storage model. An intermediate optimal amplitude was also found due to that extra HC and CO emission coming from the richer side lowered down the overall conversion efficiencies when dithering with higher amplitudes. The difference between emission data after the first catalyst and after both indicated that more catalyst capacities are critical for HC conversion.

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INTRODUCTION

Three-way catalytic converter (TWC), an indispensable device for the control of unburnt hydrocarbon (THC), carbon monoxide (CO) and nitric oxides (NO_x) emissions of gasoline engines, is a chemical reactor operating at highly transient conditions [1]. TWC can simultaneously reduce THC, CO and NO_x at certain temperature, flow rate and composition of the exhaust gas. Since three-way catalytic converter will work most effectively at stoichiometry of the combustible mixture, the majority of installation are for four-stroke, quasi-stoichiometric engines [2]. TWC is also applicable to natural gas engines, since they produce similar emission species compared to gasoline engines, except that the main THC species is CH_4 . Due to new drilling techniques and newly-found resources, natural gas (NG) is currently available at attractive prices. As such, many buses, fleets, and personal cars have been either converted to accept NG, or been replaced by NG vehicles. There are also many stationary engines already using NG as main fuel [3]. Policy-wise, as greenhouse gas regulation is currently in place in North America, controlling methane emissions from conventional gasoline engines is also of critical importance. Therefore, effective applications of TWC on natural gas engines are of high practical interests. However, the main emissions from NG SI-engine, namely CH_4 , CO and NO_x may not be as low as for gasoline SI-engines especially when the catalyst has undergone an aging period [4]. Methane production was even found over three-way

catalysts in E85-fuelled vehicles [5]. Moreover, CH_4 conversion over three-way catalyst surface is more sensitive to exhaust gas composition, i.e. CH_4 has a narrower zone of favorable oxygen level to achieve fully conversion. To improve the conversion of CH_4 , numerous possible improvements have been investigated, such as different surface metal species and arrangements [6, 7], different physical catalyst set-up [8, 9], catalyst pretreatment and preparation [10], as well as optimal engine operation techniques [11, 12, 13].

Dithering, as a term that describes oscillation of the engine charge composition related to different Lambda (normalized air fuel ratio) values, mainly creates consecutive patterns of rich/lean conditions to optimize the conversion efficiency of the catalysts. Compared to other emission reducing techniques, dithering does not affect the existing engine setup or run conditions and won't require complicated feedback control system. Therefore it is a favorable emission control solution as an exhaust retrofit. The behavior of TWC under such dynamic conditions has already intrigued many research interests in experimental demonstrations [2, 11, 12, 13, 14], numerical modeling [15, 16, 17, 18] and detailed chemistry of major emission species over noble metals (Pt, Rh, Pd) [19, 20, 21, 22]. Ishii et al. [14] showed in their experimental data that Pd-based catalysts are slightly more suitable for CH_4 conversion than Pt-based types, especially in the region of Lambda from 0.980 to 0.995 at 400 °C. Onisawa, Katsuyuki et al. [15] found that the oxygen stored in the catalyst increases with

catalytic wall temperature and lean/rich depth of the mixture. Therefore, they established an oxygen storage-release model from flow reactor experiment results of gasoline to predict the temperature and mixture composition effects. Xin et al. [22] investigated kinetics of catalytic oxidation of methane over a palladium oxide (PdO) surface by wire microcalorimetry. Their results provide valuable information regarding detailed mechanism of catalytic methane oxidation. Upon all these above researches it is still not clear that which operational parameters are the key factors for effective dithering and how they should be quantified.

The paper aims at better understanding of dithering so that dithering can be effectively applied to natural gas engines. To this end, an experimental study was conducted to explore various parameters that can be easily manipulated. Steady-state runs were first conducted for setting up references in the emission level and catalyst capacity. Next, transient runs between lean and rich engine operations were conducted to illuminate the transient characteristics of the catalyst. Dithering with certain operational parameters were also tested. Optimal operational points were found and related to the oxygen storage model from the transient runs.

EXPERIMENTAL SETUP

A single-cylinder, variable-compression-ratio Cooperative Fuel Research (CFR) engine (model F2, 1958) is used in the experiments. All the detailed engine test bench parameters are listed in Table 1 and 2. The setup of the entire test bench is schematically showed in Figure 1. The engine speed is controlled by an induction motor and a variable frequency drive which maintains the speed within a narrow window around 600 RPM. Gas injector is installed at the downstream of intake pipe for port injection of gaseous fuels, in this case NG. Fuel injection and spark time were both controlled by a MOTEC engine control system. Injection timing is set to 310 BTDC ensuring fully mixing with air. Spark timing, as set to 22 BTDC, showed stable combustion without knocking and was near maximum brake torque. The engine is naturally aspirated and no throttling or boosting was used for this testing. Compression ratio (CR) was adjusted to 6.5, in a sense that the emission will be much enough for proper catalyst study, as well as not too much avoiding risks of overheat the catalyst converter.

Table 1. Engine Test Bench Specifications

Displaced volume	612 cc
Stroke	114 mm
Bore	82.5 mm
Connecting Rod	254 mm
Number of valves	2
EVO @ 0.15 mm lift	148° CA ATDC compression
EVC @ 0.15 mm lift	-353° CA ATDC compression
IVO @ 0.15 mm lift	-343° CA ATDC compression
IVC @ 0.15 mm lift	-153° CA ATDC compression

The exhaust stream was equipped with two catalytic converters in series which allowed placement of sensors and sampling probes before, after or between the catalysts. The flow through each catalyst was restricted with a gasket with an inner diameter of 38 mm (1.5 in.). With the length of 203 mm (8 in.). This provides a volume of about 0.23 L for one catalyst, or 0.46 L for both. The catalysts are

general-purpose three-way catalysts with Palladium (Pd) and Rhodium (Rh), where the ratio is 9:1 (Pd:Rh), and a washcoat for oxygen storage. The catalysts in the current setup can be preheated and insulated so that they could be tested up to 650 C. A Horiba gas analyzer was used for measurement of NO_x , CO, HC. A wide-band oxygen sensor connected to MOTEC control system is mounted right at the front of the exhaust system (before the first catalyst), as showed in Figure 1.

Table 2. Engine operating conditions

Compression ratio	6.5 : 1
Engine speed	600 RPM
Coolant temperature	75.0 ± 1.00 °C
Intake pressure	100.0 ± 0.5 kPa
Intake temperature	28.0 ± 1 °C
Fuel	Methane
Throttle	Naturally aspiration

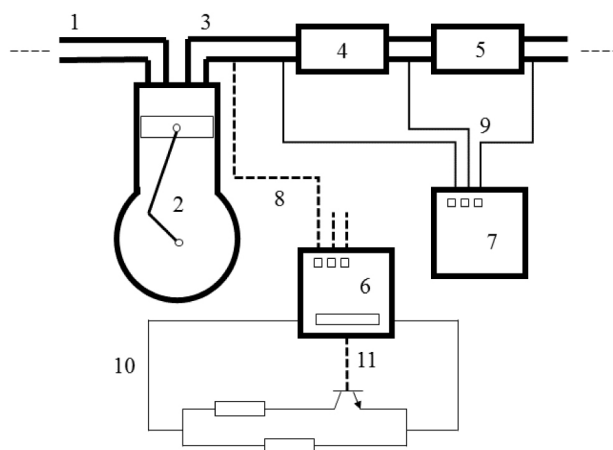


Figure 1. Schematic of Engine Test Bench: 1-intake, 2-single cylinder engine, 3-exhaust, 4-first catalyst, 5-second catalyst, 6-MOTEC system, 7-Horiba gas analyzer, 8-oxygen sensor signal, 9-exhaust sampling probes, 10-TVP measuring circuit, 11-dithering control voltage signal.

Dithering was achieved by the following control strategy: The amount of fuel injected for each cycle is controlled by MOTEC system, referring to a fuel mapping table with regard to engine speed and throttling valve positioning (TVP). To be more specific, MOTEC will receive both engine speed signal and TVP signal and then determine the injected fuel amount by reading the value from the corresponding location in the mapping table. In the current setup, throttling valve was took off from the engine to enable naturally aspirating air-intake mode. Instead, two parallel resistors were connected to throttling valve positioning measuring system, as showed in Figure 1, while a transistor was also connected to one of the two resistors. Therefore, by dynamically controlling the transistor base voltage, the measuring circuit can switch between measuring one resistor or two parallel resistors, to fake as two individual TVPs. In this way, MOTEC system can switch between two locations in the fuel mapping table back and forth, and dither the injected fuel amount accordingly. By setting a constant value to the input voltage signal, one can run constant Lambda engine operation, which refer to steady-state runs. By setting a wave function with very low frequency to the input voltage signal, one can run transient engine operation, rich and lean. By changing the frequency of input voltage signal, this

setup can enable dithering at different frequencies. By writing the values in the fuel mapping table, it can enable steady-state runs at different Lambda values, transient runs between different lean and rich conditions, as well as dithering at different Lambda midpoints and amplitudes.

RESULTS AND DISCUSSION

Steady State Runs

This section shows the results of runs with steady Lambda values, to identify general emission levels of this specific engine and corresponding catalyst capacities. All the data points (except data point Lambda = 0.995 in Figure 3) are the average values of emission at steady states around 1000 cycles (approximately 3 minutes), which reasonably represent the catalyst steady-state performances.

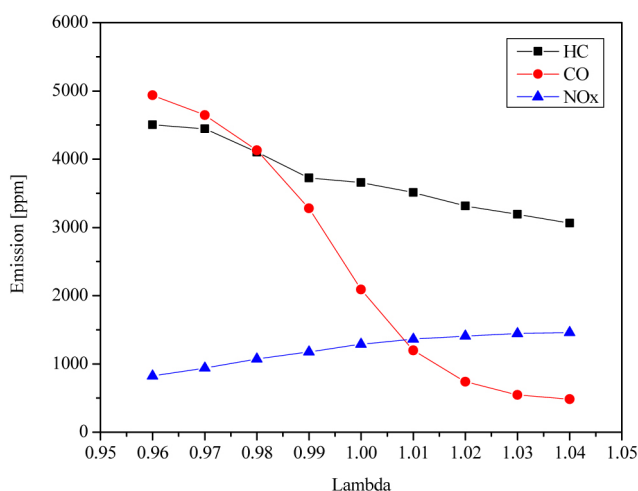


Figure 2. Experimental engine-out emissions at different Lambda values

Figure 2 shows the experimental results of emissions in the exhaust mixture before the catalyst, i.e., sampled emission from the first sampling position in the exhaust system. CO emission decreased and NO_x emission increased when the engine charge shift from rich to lean (Lambda increases), which matches the fundamental combustion theory [23]. Although HC emission, mostly CH₄, also slightly decreased when engine ran leaner, the overall level of HC emission is still relatively high compared to normal gasoline vehicle emissions. That is due to the nature of stable molecular structure of CH₄ molecules, which makes CH₄ a difficult fuel to burn. If the engine cylinder has large crevice volumes or high crevice/clearance ratio, which is the situation of the CFR engine, certain amount of CH₄ will be squeezed into crevices, left unburnt during compression stroke, come out crevices during expansion stroke and exhaust as emissions. Therefore, this scenario of above experimental emission level will refer to natural gas engines with large crevices or crevice/clearance ratios, especially aged ones with extra crevices near piston rings. Also it could represent some large stationary engines of syngas, biogas, etc., which complete combustion is hard to achieve.

Figure 3 shows the experimental results of emissions after the two catalysts. CO emission was almost zero on the lean side while NO_x was zero on the rich side, due to their chemical essence of reducer and oxidizer. What triggered interests is that HC conversion is only favorable at slight rich engine mixture, where Lambda ~ 0.995. On

either side of the favorable point, HC conversion is unacceptably lower. It was also observed in studies from other researcher. [14, 24] This phenomenon is often explained by the oxygen storage nature: on the rich side, unburnt HC do not have enough oxidizer to react with and be reduced, while on the lean side, excess oxygen molecules occupied most surface sites so that HC molecules cannot even attach to the catalytic surface. Therefore HC were left on both sides. For the data point of Lambda 0.995 showed in Figure 3, it was controlled manually by the researchers between 0.99 and 1.00. The result is only the average emission values over 100 cycles of 20s. If the Lambda value stayed on 0.99 or 1.00 for relatively long time, HC conversion will decrease dramatically. Thus although favorable HC conversion can be achieved in the steady-state runs, it is still very hard to maintain for certainly steady operations. Slight shift of Lambda will cause huge emission difference. The following sections tried to, on one hand, disclose how catalysts performed in the transient runs which are common engine operations; on the other hand, come up with a strategy to achieve high conversion efficiencies for all three major emissions. As expected, dithering with certain parameters would show its potential being more stable for favorable emission conversion.

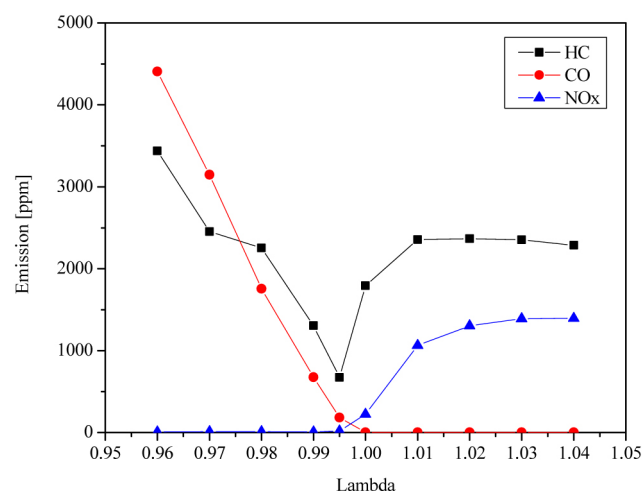


Figure 3. Experimental catalyst-out emissions at different Lambda values

Transient Runs between Steady States

Transient runs between steady lean and rich states were experimentally performed to explore how all three emission species react to varying Lambda (i.e. oxygen levels in the oxygen storage model discussed later).

Figure 4 shows transition from slightly lean steady state (Lambda 1.00-1.01) to rich state (Lambda 0.97-0.98) and Figure 5 shows the reverse transition. Note that an optimal operation zone was only found during the transition from lean to rich, where three emission species are all low (circled out in Figure 4), but not the reverse transition. An oxygen storage model was proposed to explain these phenomenon: oxygen is largely pre-stored on the catalyst surface when running lean. When rich exhaust gas reaches the catalyst, unburnt hydrocarbon (HC) and carbon monoxide (CO) try to attach the catalyst surface. In the meanwhile, oxygen starts to diffuse back to the gas-phase which release active sites on surface and enable HC and CO to attach and react with surface oxygen. NO_x also starts to attach to the surface and reacts as an oxidizer. Therefore all three