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DOI:
10.1016/j.jece.2016.04.015
10.1016/j.jece.2016.04.015

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Document Version
Peer reviewed version

Citation for published version (Harvard):
Hasan, AO, Abu-Jrai, A, Al-Muhtaseb, AH, Tsolakis, A & Xu, H 2016, 'HC, CO and NOx emissions reduction efficiency of a prototype catalyst in gasoline bi-mode SI/HCCI engine', Journal of Environmental Chemical Engineering, vol. 4, no. 2, pp. 2410–2416. https://doi.org/10.1016/j.jece.2016.04.015, https://doi.org/10.1016/j.jece.2016.04.015

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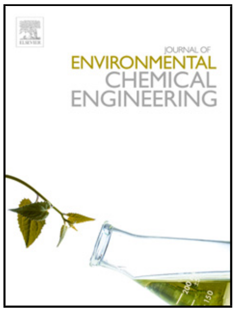

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PII: S2213-3437(16)30145-2
DOI: http://dx.doi.org/doi:10.1016/j.jece.2016.04.015
Reference: JECE 1068

To appear in:

Received date: 30-1-2016
Revised date: 22-3-2016
Accepted date: 12-4-2016

Please cite this article as: Ahmad O.Hasan, Ahmad Abu-Jrai, Ala'a H.Al-Muhtaseb, Athanasios Tsolakis, Hongming Xu, HC, CO and NOx emissions reduction efficiency of a prototype catalyst in gasoline bi-mode SI/HCCI engine, Journal of Environmental Chemical Engineering http://dx.doi.org/10.1016/j.jece.2016.04.015

HC, CO and NO_x emissions reduction efficiency of a prototype catalyst in gasoline bi-mode SI/HCCI engine

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Research Highlights:

1- Emissions under lean and stoichiometric HCCI/SI engine operation have been studied.
2- HC and CO emissions reduction was in the range of 90- 95%.
3- NO_x reduction under lean engine operating conditions was in the range of 35 to 55%.
4- HC, CO in HCCI mode are heavily dependent on engine load regardless of A/F ratio.
5- Catalyst efficiency in reducing emissions is related to the exhaust gas conditions.

Abstract

Lean and highly diluted homogeneous charge compression ignition (HCCI) engines offer great potential in improving vehicle fuel economy and contribute in reducing CO_2 emissions. Hydrocarbons and CO emissions from HCCI engines can be higher than those from spark ignition (SI) engines, especially at low engine load when the residual gas required to control NO_x emission are elevated. Although, NO_x emissions are significantly low, a bi-functional after treatment device will be required to control HC, CO and NO_x emissions under lean and stoichiometric (oxygen free) engine operating conditions. This paper describes studies on the NO_x, HC and CO emissions reduction efficiency of a prototype catalyst, under lean and stoichiometric engine conditions at different loads, speeds, and A/F. A comparative study of catalyst performance will be analysed under HCCI stoichiometric and SI operation under three engine speeds and load of 4 bar (Net Mean Effective Pressure, NMEP). Experimental results indicate that the HC and CO emissions reduction over the prototype catalyst was in the range of 90- 95% while the maximum NO_x emissions reduction under lean engine operating conditions was in the range of 35% to 55%. The catalyst efficiency in reducing the three pollutant emissions is closely related to the exhaust gas conditions (e.g. temperature and space velocity), oxygen and composition i.e. NO_x, CO and HC concentrations.

Abbreviations: BC: Before catalyst; AC: After catalyst; THC: Total hydrocarbon; CO_2: Carbon dioxide; CO: Carbon monoxide; NOx: Nitrogen oxide; HCCI: Homogeneous charge compression ignition; A/F: Air fuel ratio; NMEP: Net Mean Effective Pressure; SI: Spark ignition; CI: Compression ignition; TWC: Three-way catalytic converters; Pt/Rh: Platinum /

radium; GDI: Gasoline direct injection; VCT: Variable cam timing; CPS: Cam profile switching; λ : Lambda.

Keywords: Emissions; Combustion; Catalyst; HCCI engine; Three-way catalytic converters.

1- Introduction

Homogeneous charge compression ignition (HCCI) engine takes premixed air/fuel mixture as in conventional spark ignition (SI) engines and ignites the mixture by compressing it in the cylinder without a spark plug, as in conventional compression ignition (CI) engines Lean burn combustion is achieved through homogeneous mixture formation and compression ignition enabling combustion temperature much lower than that of conventional SI and CI engines. Due to the lean mixture low temperature combustion, nitrogen oxides (NOx) emissions are reduced dramatically and fuel economy is improved. [1-6]. However, greater amounts of hydrocarbon (HC) and carbon monoxide (CO) emissions are released relative to conventional SI and CI engines. The oxidation reactions of HC and CO emissions during the expansion stroke are reduced due to the lower combustion temperature. Unlike the gasoline spark ignition gas engine or diesel engine, the homogenous charge compression ignition (HCCI) produces a low-temperature, flameless release of energy throughout most of the entire combustion chamber. The fuel mixture existed in the combustion chamber are burned simultaneously. These phenomena are able to produce power similar to conventional gas engines, with less fuel needed to do it. Heat is a necessity for the HCCI process to take place, so a traditional spark ignition (SI) mode is needed to be used when the engine is started cold to generate heat inside the cylinders and rapidly heat up the exhaust catalyst and enable HCCI engine operation. HCCI operation builds on the integration of other advanced engine technologies – some of these are already have been in production and can be used to existing gas engines.

Even if HCCI operation at high loads is obtained, the NO_x reduction benefit is small compared with conventional SI engine with three-way catalytic converters (TWC). Meanwhile, the fuel consumption advantage of gasoline HCCI over SI combustion is reduced at high load due to the reduced level of throttling. For high-load operation, it may require to again switch to traditional SI or CI operation. In HCCI engines the ignition timing is subject to chemical kinetics of the reactants and dependent on the fuel composition and parameters such as, air/fuel ratio, and the thermodynamic state of mixture [7-9].

Hydrocarbon and carbon monoxide levels of HCCI engines vary between experimental conditions (e.g. engine technology and combustion modes. Though they are found to be similar or higher than those of SI engines and both CO and HC emissions are increase at light loads. Although, TWC technology has been perfected over the years for use in SI stoichiometric combustion engines, under HCCI operation or lean operation those catalysts are not effective in reducing NO_x emissions. In addition switching engine operation to lean, stoichiometric or even rich is making the application of conventional oxidation catalysts challenging especially at low exhaust temperatures associated with HCCI engines [10-12].

Significant advances have been made in improving TWC performance, resulted in faster light off, higher conversion efficiency, and greater thermal durability. Besides, improvements in fuel quality and engine management led to tighter emissions standards being achievable, concomitant with reducing the cost by lowering precious metal loadings [13-15]. The Pt/Rh catalyst with single-layer wash coat has poor HC reduction under lean A/F operation, but if the two precious metals are separated by duel layers, the HC-reduction will be substantially improved. In addition, the drop in HC reduction efficiency during the transient phase of lean A/F to stoichiometric can be substantially minimized by optimizing the loading of Pt and Rh contents in the wash coats [16, 17].

Manoj et al [18] have used cordierite/Pt catalyst on bio-diesel single cylinder engine and found that, by increasing engine load, the NOx conversion has been reduced dramatically due to ammonia desorption and increased emission of unburned hydrocarbons. Annaprabha et al [19] have tested a selective catalytic reduction catalyst for NOx from diesel engine, with low concentration; they reported that, CeCac (with highest cerium surface weight percent) achieved the highest reduction efficiency over all temperature. Woojoon et al [20] presented V2 O5 – Ce O2/CVC-TIO2 Catalyst on the NH3-SCR and DeNOx with/without SO2, and water vapour process, the catalyst showed good SCR activity for NOx removal and exhibited enhanced SCR activity in the presence of SO2 and water vapour at low temperature.

This manuscript presents a study of designing automotive three-zone prototype catalytic converter for the effective control of NO (Nitric Oxide), HC (Hydrocarbon) and CO (Carbon Monoxide) emissions under both lean and stoichiometric engine operation. The work was carried out using V6 engine operating in SI and HCCI mode using commercial unleaded gasoline fuel. The catalyst efficiency in reducing the three pollutant emissions is closely related to the exhaust gas conditions (e.g. temperature and space velocity), oxygen content and composition i.e. NO_x, CO and HC concentrations, also the effects of engine load, and air/fuel ratio on exhaust emissions and fuel consumption were explored, for both SI and HCCI combustion modes.

2- Experimental

2.1 Engine -The experimental work was performed on a V6 HCCI / SI mode gasoline direct injection (GDI) engine Figure (1).The engine intake and exhaust camshafts were built with variable cam timing (VCT) and cam profile switching (CPS) system. Fuel direct injection pulse width is adjusted by the engine management system to maintain the required value of λ. The engine was coupled to an eddy-current dynamometer for speed and load control, twelve thermocouples type K were installed to monitor the exhaust and inlet temperature. Intake and exhaust cam timing for minimum residuals were chosen to start the engine with spark ignition combustion. When engine block achieved steady condition of temp level in SI mode, the HCCI combustion mode was started. Negative valve overlap was used to increase the amount of exhaust gas retained in the cylinder to achieve HCCI combustion. The temperature of the intake air was controlled by a thermal management system. Fuel flow was measured by an AVL gravimetric meter, and the fuel direct injection pulse-width was adjusted by the engine management system to maintain the required air/fuel ratio. The engine was connected to an EC38 eddy current dynamometer. A DSPACE-based system coupled to a computer using MATLAB/SIMULINK software is used to control the engine parameters during operation and to record engine data. Details of the engine specification are listed in Table (1).

***2.2 Catalyst* -** The prototype three zones monolith catalyst, with optimized order of the zones, was supplied by Johnson Matthey (UK) and was connected to the actual engine exhaust manifold (Figure 1). The first zone was designed to reduce HC and NOx under lean and stoichiometric engine conditions at high temperatures >400°C, the second zone was designed to reduce NOx by reaction with hydrocarbon under lean engine operation in the temperature range of 250°C –

400°C. Both zones contain non-precious metal catalyst supported on alumina or zeolite. The third catalyst zone (Pt based) was designed to control part of the exhaust hydrocarbons and CO at temperatures below 300°C.

2.3 Emissions analysis - A Horiba MEXA 7100 DEGR equipped with a heated intake line was used to measure total hydrocarbons, carbon monoxide, carbon dioxide, NOx and oxygen..The heated line and pre-filter are maintained at 190ºC. The analyzer equipped with flame ionization detector (FID) was used to measure total hydrocarbon in ppm methane (CH_4) equivalent. The measurement of HC represents wet HC concentration. Carbon monoxide (CO) and carbon dioxide (CO_2) were analysed by non-dispersive infrared (NDIR) detector and NOx by a chemiluminescene detector (ClD). The λ (air/fuel ratio relative to stoichiometric), temperature and pressure of the exhaust gases at the engine exhaust manifold were also measured. The system should be calibrated using suitable bottled span and zero calibration gases.

2.4 Experimental procedure - The tests have been divided in two load groups, low load and higher load, three air-fuel ratios (represented by λ) as well as three engine speeds were used in this study (see Table 2 and Table 3). This approach allows consideration of varied parameters (e.g. space velocity, temperature) on the catalyst efficiency in reducing regulated emissions.

2.5 Engine Data processing- The data processing procedure involves averaging over 100 consecutive pressure cycles (including pressures other than in-cylinder), temperatures, fuel consumption and other parameters. Parameters such as: NMEP, rate of pressure rise, COV of NMEP and fuel consumption were calculated in the SIMULINK model; only an averaged value was calculated during data processing. All data processing has been accomplished using MATLAB software. Since data was acquired with quite a large variation of ambient conditions, standardization to normal conditions was performed.

3- Results and Discussion

3.1 HCCI operation, effect of engine loads

Hydrocarbon and CO emissions are influenced by the in-cylinder conditions (i.e. temperature) and the homogeneity of the fuel with air. Increasing engine load from 3-4 bar or 4-5 bar NMEP (Net Mean Effective Pressure) improves HC & CO oxidation regardless of the air fuel ratio (Figures 2-5) which leads to significant reduction of HC and CO emissions, on the other hand the opposite trend seen in the case of NO_x emission when the engine load was increased. In case of HC this was presumably because crevice loading of HC with lower load has a bigger effect on HC emissions than that with a higher load. The burned gas temperature at low load 340°C (Table 2) seems to be too low for the crevice hydrocarbons to be oxidized during the expansion stroke, comparing to the higher gas temperature of 420°C presented at higher load Table (3). The lower gas temperature is not favourable for HC oxidation in the exhaust ports. Marriott et al.[21] performed cycle-resolved HC exhaust emissions. They found that HC emissions were decreased with increased load and/or fuel stratification. They also observed two different mechanisms responsible for HC reduction. At higher load the combustion temperature is higher and promotes complete oxidation. When higher stratification was applied two factors were presented. Firstly, increased stratification means higher fuel concentration in the bulk-gas, therefore higher combustion temperatures and complete combustion is promoted. Secondly, increased stratification leads to reduced amounts of fuel in the quenching zones. Furthermore, the low combustion gas temperature presented at low load can result in incompletion of the combustion process and cause higher HC Maurya et al. [22]. Similarly when load is decreased CO emissions increased. This rapid increase resulted from the quenching of the CO oxidation process in the lower load as the gas temperature dropped. Dec et al. [23] predicted that for low loads, incomplete bulk-gas reactions should play a significant and perhaps dominant role in CO emissions and this contributes to HC emissions. NO_x reduction has been a result of lower combustion temperature in the homogeneous combustion associated with dilution and an increased heat capacity as an exhaust gas is used to dilute the cylinder mixture.

3.2 HCCI operation, effect of air fuel ratio

Allowing increasing air-to-fuel ratio has been considered as one of the advantages of HCCI combustion. As air-fuel ratio (λ) increased from 1.2-1.6 at 4 bar NMEP (leaner operation) (Figures 3 and 4) a more diluted in-cylinder mixture leads to lower temperature and therefore lower NO_x emissions, on the other hand when (λ) has been increased further the fresh charge becomes colder which in turn deteriorates auto-ignition and resulted to higher HC & CO emissions. Another effect has influenced the emissions is that; the total heat release and average combustion temperature are reduced as the fuel rate reduced. As a result, fuel/air mixtures are subject to low combustion and post-oxidation temperature and less complete oxidation of HC and CO to CO_2. This is consistent with CO and THC emission results reported by Dec et al. [24].

3.3 HCCI operation, effect of engine speeds

Increased engine speed reduces engine out HCs but does not influence significantly CO emissions Figure (6) and (7) this fact has been ascribed to thermal conditions at which the HCCI engine operates. Increasing engine speed increases heat flux from the engine; therefore air charge has been warmer and in-cylinder temperature at the end of compression stroke has been higher. Furthermore, combustion efficiency has increased and as a result, HC emissions have decreased. On the other hand NO_x emissions tend to be reduced with increasing engine speed, this is mainly due to an improved mixing process that in turn improves charge homogeneity and reduces NO_x emissions. HCCI stoichiometric and SI operation, engine load 4bar NMEP, $\lambda = 1$. HCCI stoichiometric, SI mode were conducted under similar engine operation conditions of 4bar NMEP with three different engine speeds Table (4) Comparisons are drawn between both combustion modes; focus mainly on, a) the catalytic conversion of HC, CO and NO_x emissions b) engine fuel consumption c) exhaust gas temperature.

The in-cylinder conditions (i.e. temperature) and oxygen availability have a significant effect on the fuel combustion. This picture is very clear under similar engine conditions. HCCI stoichiometric and SI mode (Figure 8) shows unburned hydrocarbons at exhaust gas temperature of 444°C were much higher from HCCI against SI combustion at 670°C (Table 4). This difference in HC emissions is strongly affected by the engine operation modes due to the higher exhaust gas temperature in SI engine operation. On the other hand, the opposite trend seen in the case of NO_x emission in the SI operation mode (Figure 9).

Figure (10) shows that the HCCI combustion with residual gas trapping produced much less CO emissions than SI engine mode which was confirmed by other studies using the residual gas trapping method [25]. The reduction in CO emission is likely caused by the recycling of burned gases and their subsequent conversion in to CO_2 in the next cycle. It should be noted that HCCI combustion in the 4-stroke gasoline engine had been always associated with higher CO emissions than the SI combustion until the residual gas trapping method was employed [26].

The main benefits of the HCCI engine operation is the savings in fuel consumption (g/hr). From Table (4) it can be observed that under the same engine load 4 bar NMEP (i.e. same power) the fuel consumption for HCCI operation was approximately up to 14% lower when compared to SI engine operation. This is mainly a result of the reduced pumping losses due to fully open throttle operation.

3.4 Catalyst efficiency

Under variable engine conditions prototype catalyst convertor covered a wide range of engine operations in both modes, even when changing air fuel ratio (e.g. shifting the engine operation to leaner combustion 1.2-1.6), and shifting to higher load (e.g. 3-5 bar) respectively does not influence the engine exhaust gas temperature significantly this led to approval consistency of HC & CO conversions about 90% over the catalyst independently from in-cylinder conditions. This is the effect of interference of three parameters: retention time (space velocity), exhaust temperature, and exhaust gas composition (HC, CO, and CO_2 due to high heat capacity).

The most important factor toward CO conversions correlated to the retention time (space velocity-engine flow rate) – the highest conversion up to 92% (Figure 2) was achieved when engine flow rate is low at space velocity of 23 kh^{-1} (Table 2). Even at higher exhaust gas temperatures the increase in the engine load can not cover the increase in the flow rate and hence, conversion efficiency is generally lower. The best HC conversion was 96% presented at $\lambda = 1.4$ with engine speed of 2000 rpm for both engine loads. However, the prototype catalyst has shown excellent HC & CO conversion regardless of the operation modes and exhaust gas temperatures, these results have been led to a less HC difference present after the catalytic convertor between the two modes.

Minimal catalytic conversion of NO_x emissions up to 55% efficiency occurred at HCCI mode this is correlated with CO and HC emissions. When CO and HC emissions are relatively low (and exhaust temperature is low) the CO oxidation deteriorates and allows higher efficiency of NO_x reduction (more CO available for reduction reaction). There is a possibility of similar effect with H_2 in the exhaust gas. An optimal flow rate required for the best NO_x conversion, in this case, the most effective flow rate oscillates around space velocity of 28 kh^{-1}. Despite the catalyst lower NO_x conversion efficiencies under HCCI mode, NO_x emissions after the catalyst were kept at lower values compared to SI.

Conclusions:

(i) Engine-out emissions: Hydrocarbon, CO and NO_x are influenced by engine operation and combustion mode (i.e. HCCI or SI). HC and CO emissions were higher in HCCI mode while NO_x emission is found to be more in SI. Analysis shows HC, CO in HCCI mode are heavily dependent on engine load regardless of air fuel ratio, not significant effect of engine speed on CO emission as it was on HC emissions. Fuel saving up to 15% is found in the HCCI mode due to fully open throttle operation.

(ii) Post-catalyst emissions: High consistency of HC & CO conversion was approximately 90% over the prototype catalyst independent from in-cylinder conditions which has been designed to combine the catalytic functions required to oxidise CO and hydrocarbons, while simultaneously reducing NO_x over the expected range of exhaust-gas temperatures and stoichiometries during operation of an engine in both HCCI and SI modes. During lean HCCI operation, this catalyst shows more efficient conversion of HC, CO than NO_x conversion which was about 90% and 54% respectively.

(iii) Retention time (Space Velocity) one of the major factor affected CO conversion, conversion was achieved of 92% at lowest space velocity (23 kh^{-1}), An interesting results related to retention time found that the optimum catalytic conversion of the three compounds occurred together (HC, CO, and NO_x) presented at load of 3 bar, $\lambda = 1.4$ with catalytic efficiency of 96, 91, and 53% respectively which is oscillates around (28 kh^{-1}).

Acknowledgements: The authors would like to thank Johnson Matthey Plc and Shell Global Solutions UK for their support for this work.

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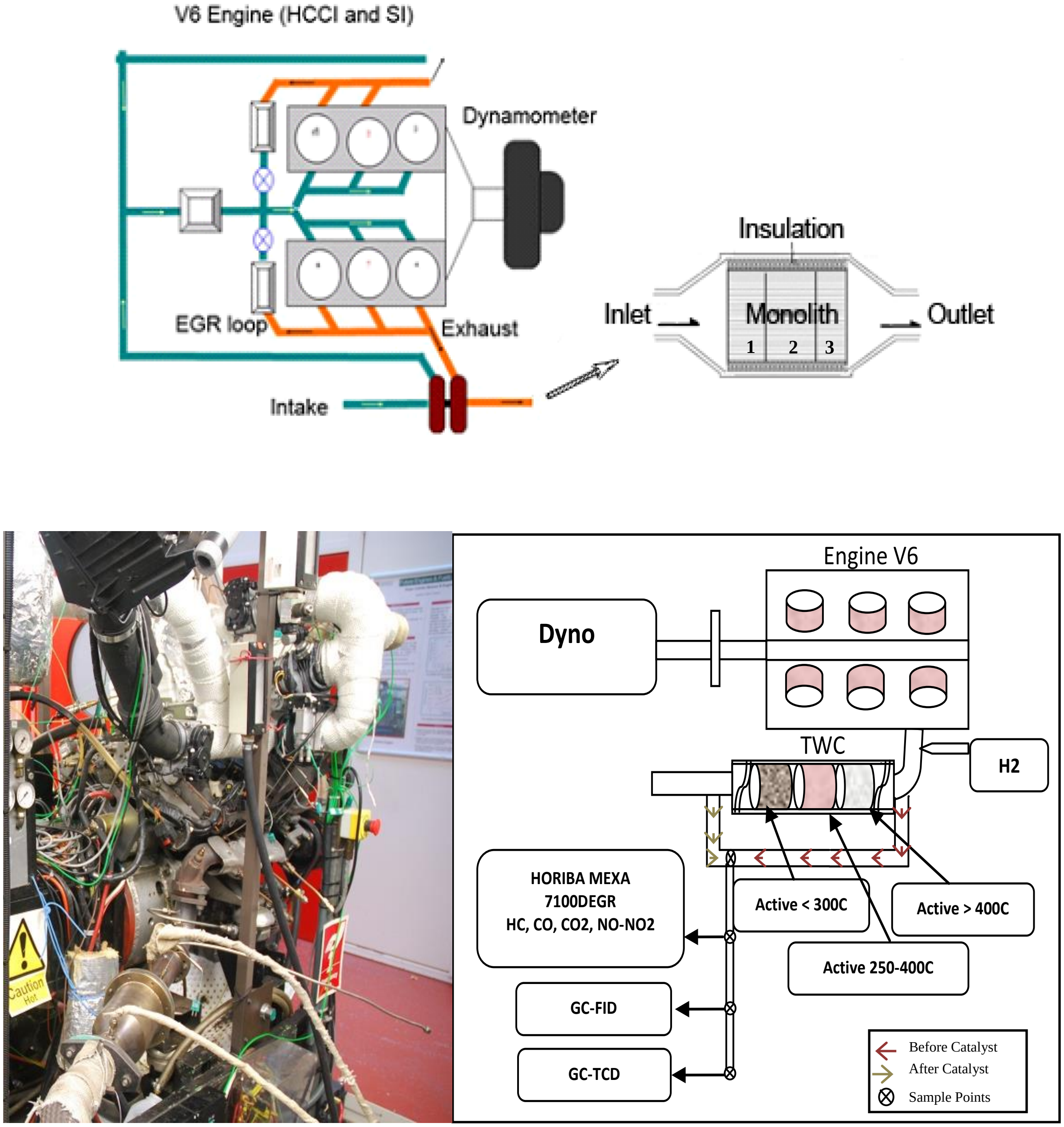

Figure 1: Schematic diagram of a V6 engine system and the prototype catalytic converter.

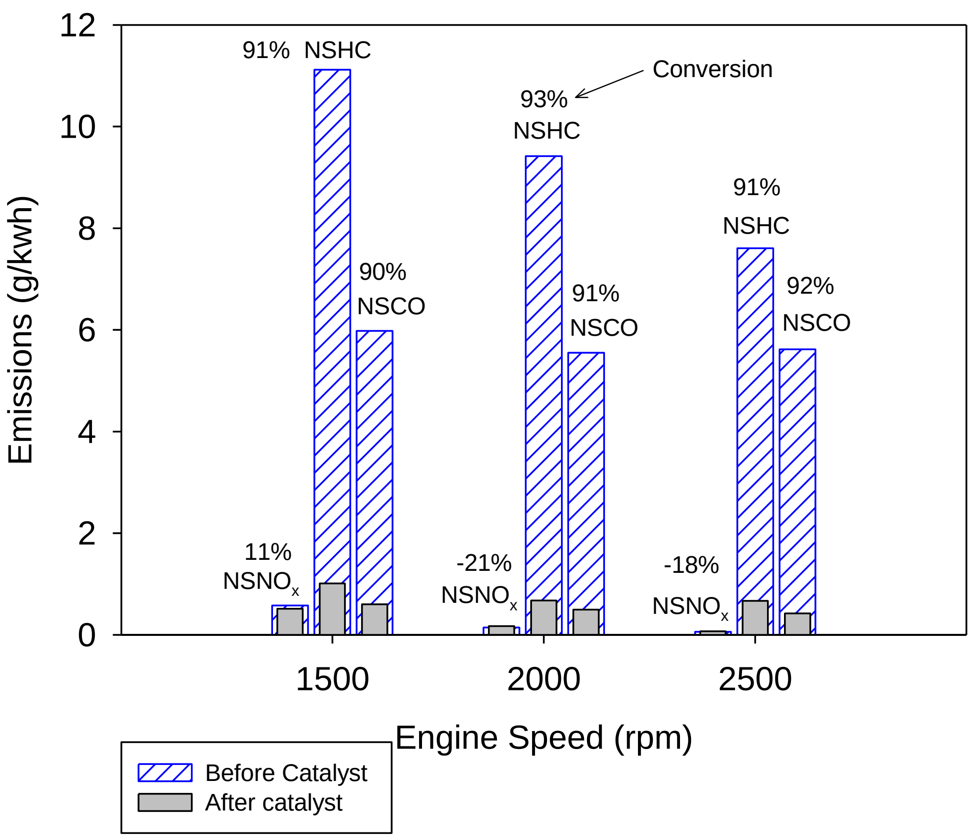

Figure 2: Catalytic conversion efficiency of HC, CO, and NO$_x$ (3bar NMEP), HCCI, $\lambda = 1.2$.

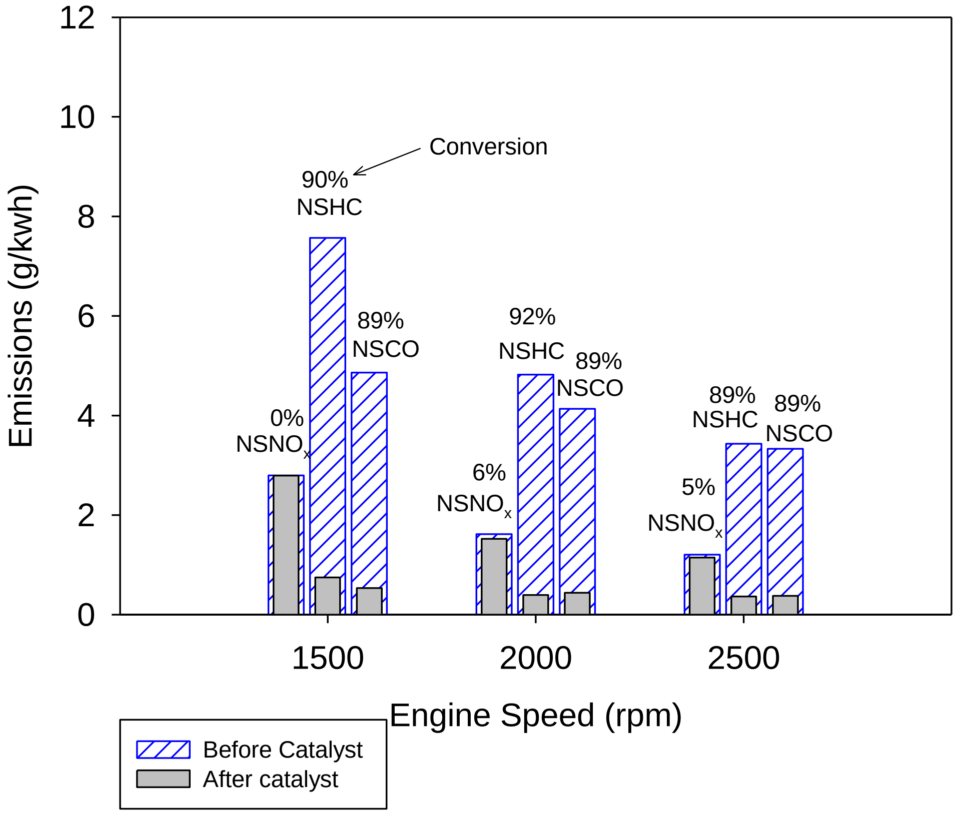

Figure 3: Catalytic conversion efficiency of HC, CO, and NO_x (4bar NMEP), HCCI, $\lambda = 1.2$.

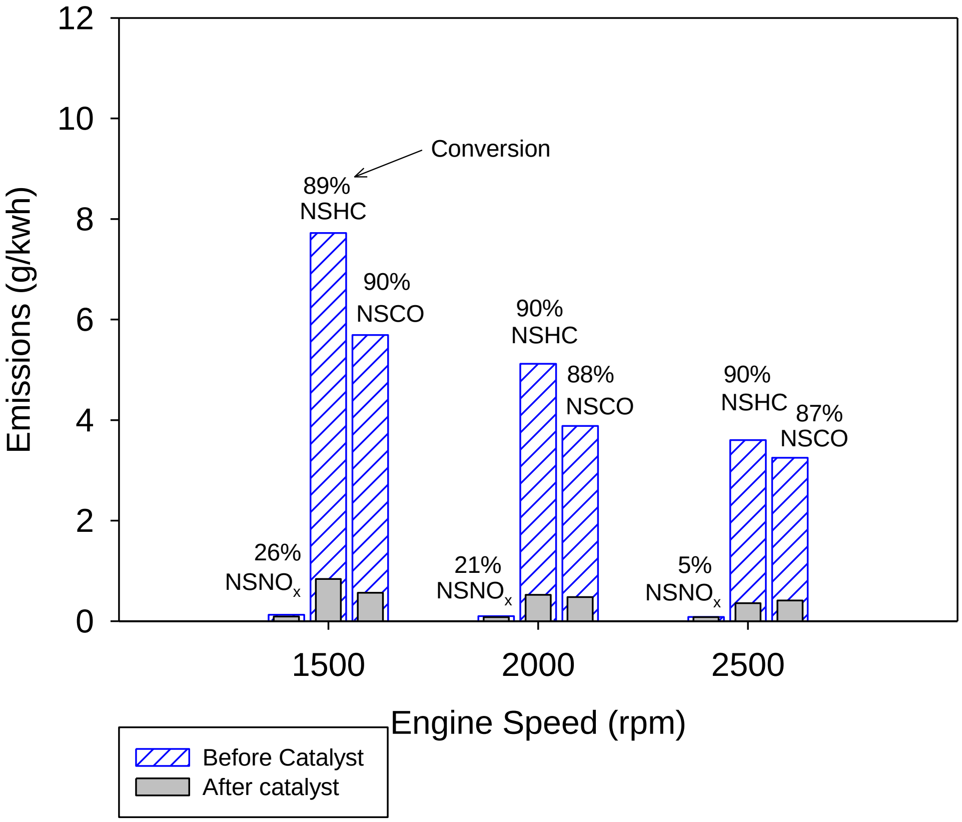

Figure 4: Catalytic conversion efficiency of HC, CO, and NO_x (4bar NMEP), HCCI, $\lambda = 1.6$.

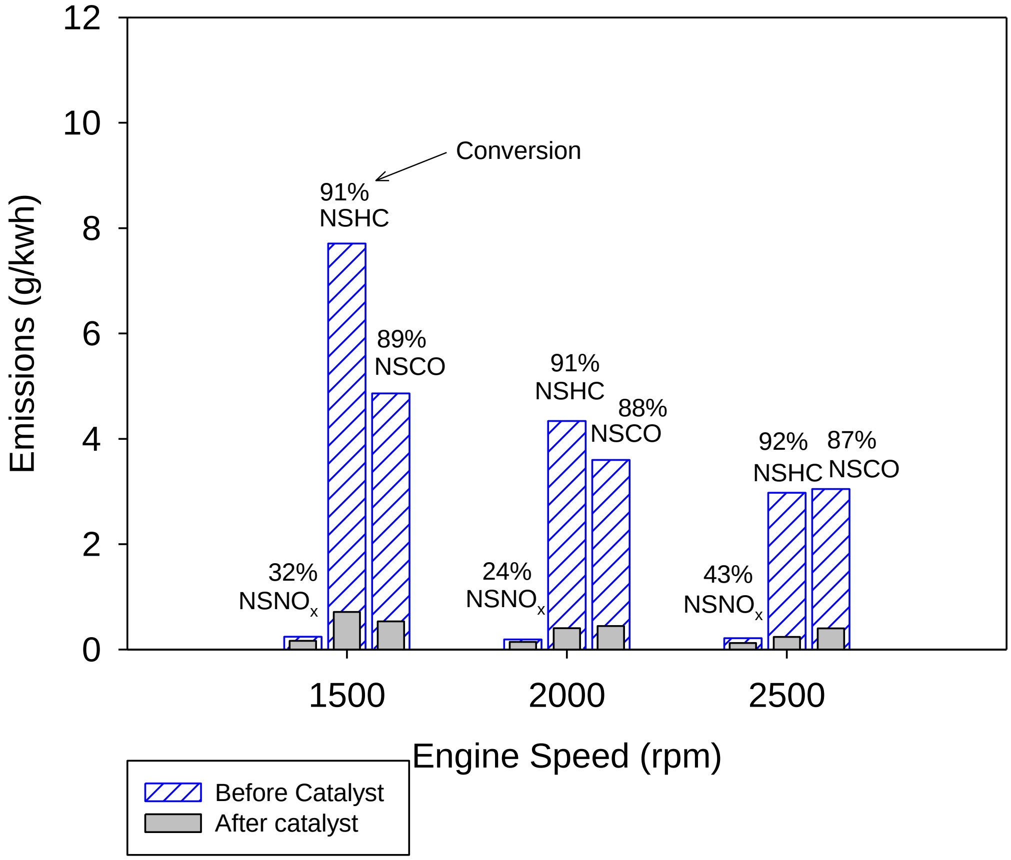

Figure 5: Catalytic conversion efficiency of HC, CO, and NO_x (5bar NMEP), HCCI, $\lambda = 1.6$.

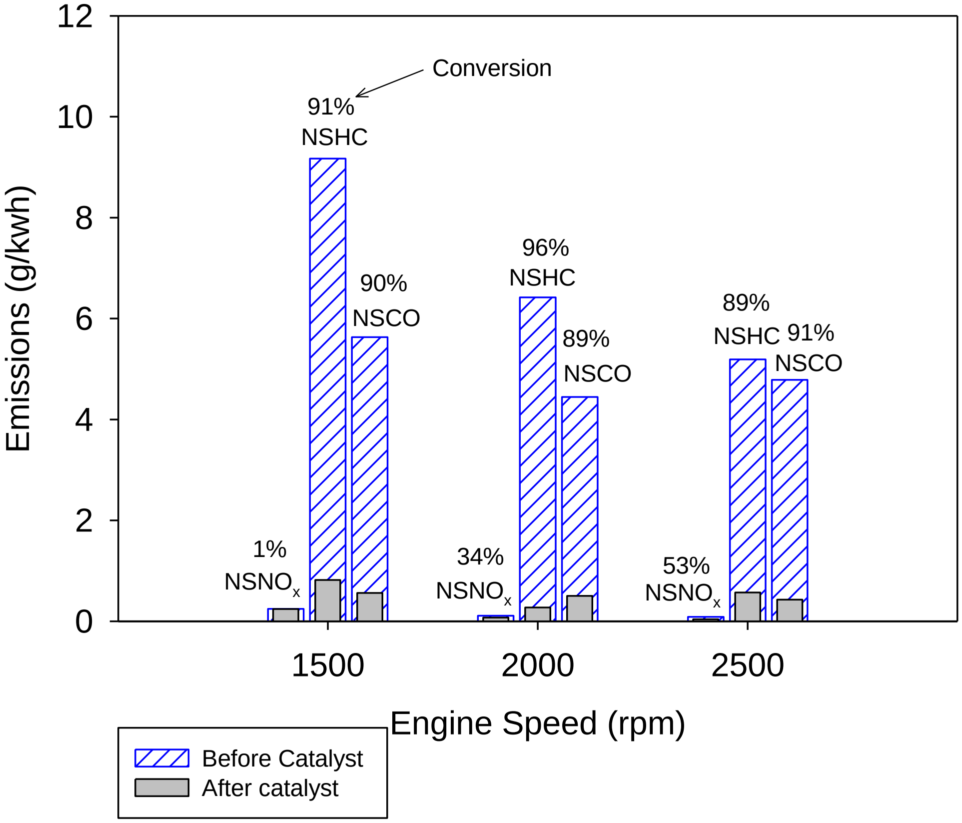

Figure 6: Catalytic conversion efficiency of HC, CO, and NO_x (3.5bar NMEP), HCCI, $\lambda = 1.4$.

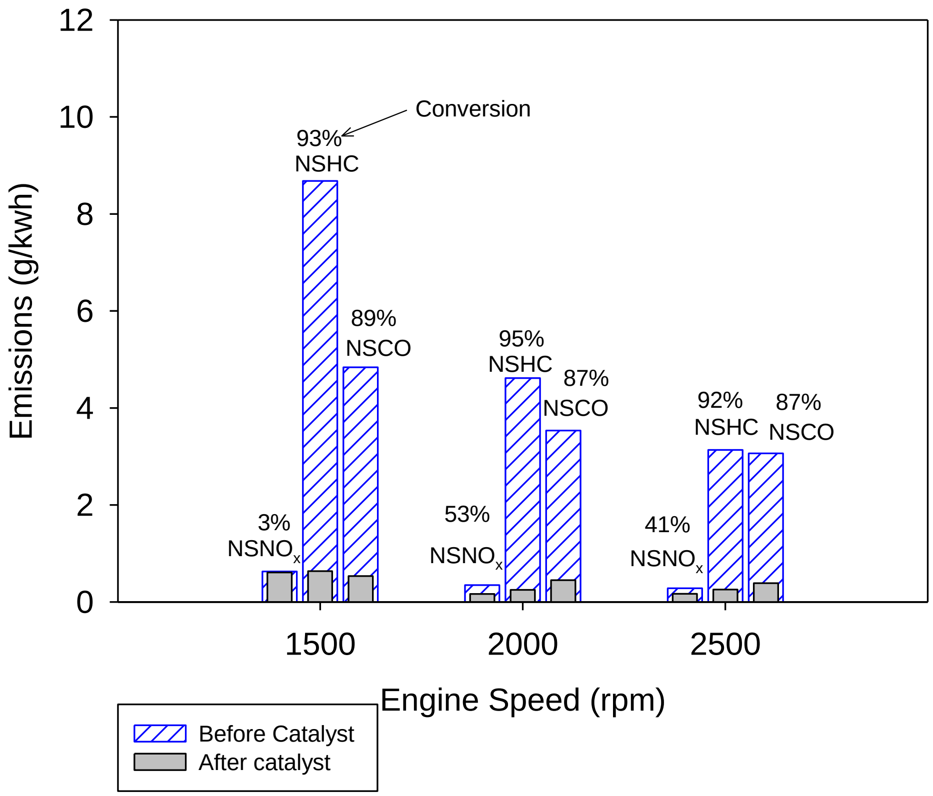

Figure 7: Catalytic conversion efficiency of HC, CO, and NO_x (4.5bar NMEP), HCCI, λ = 1.4.

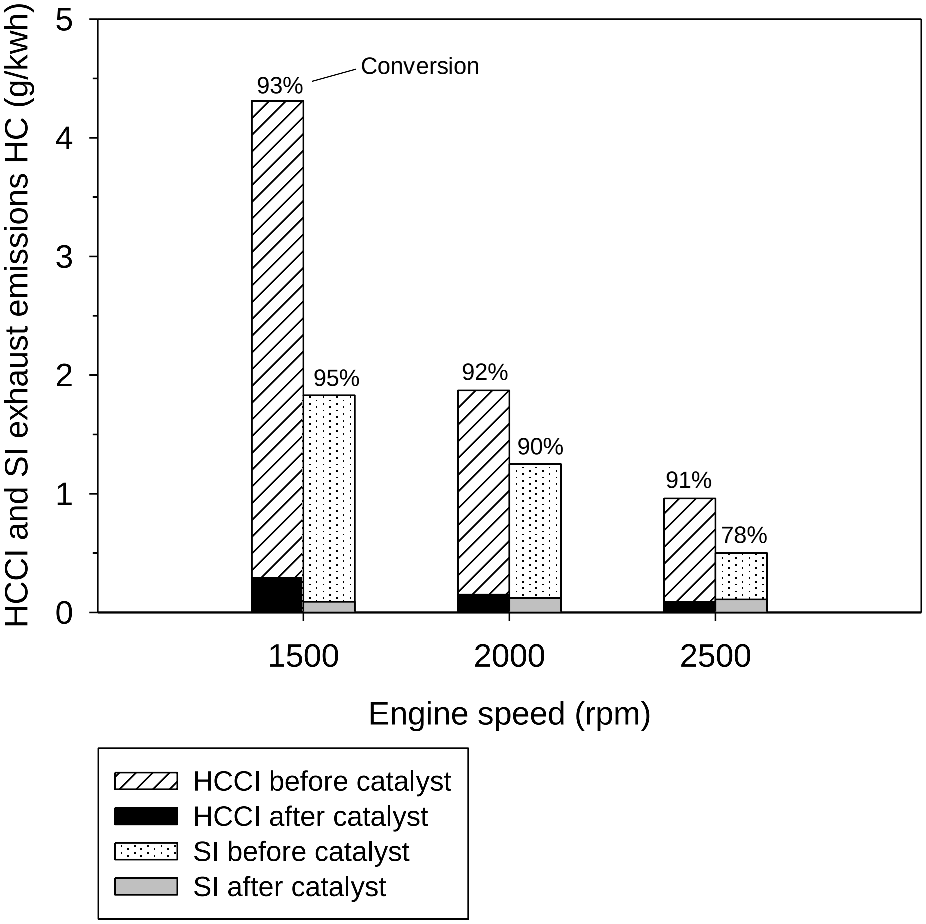

Figure 8: Catalytic conversion efficiency of HC, (4bar NMEP), HCCI, SI, λ = 1.

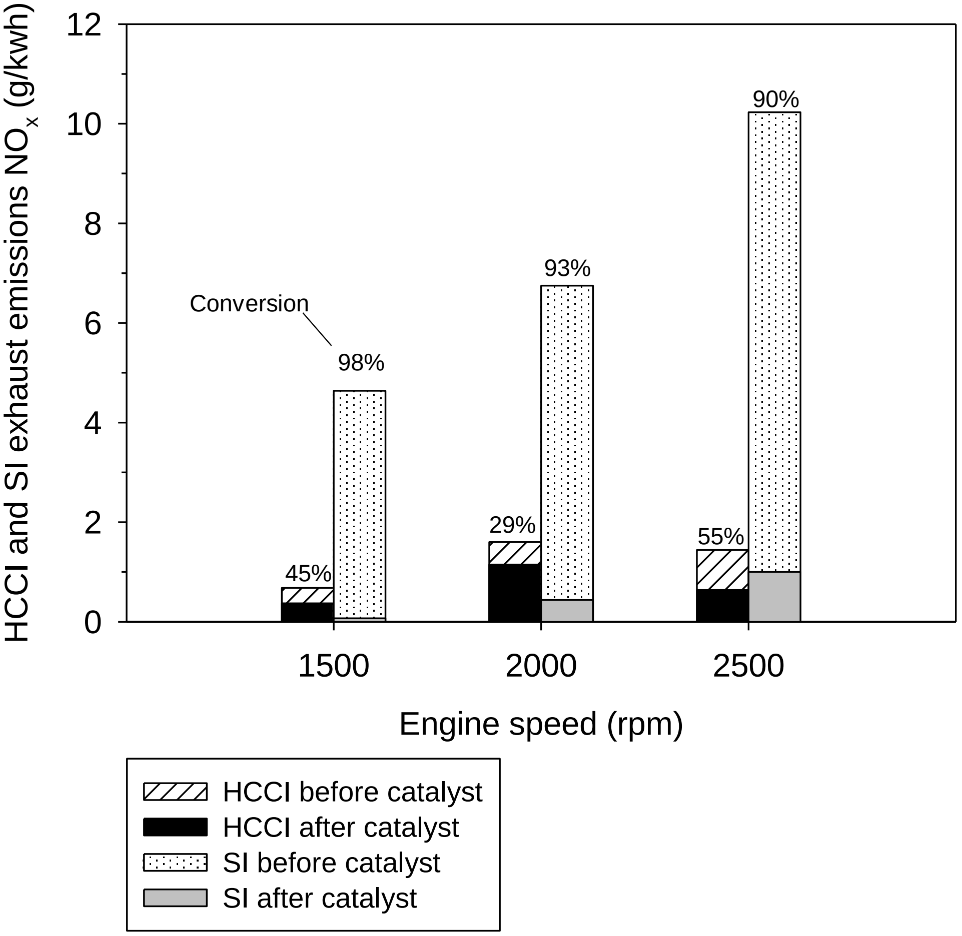

Figure 9: Catalytic conversion efficiency of NO$_x$, (4bar NMEP), HCCI, SI, $\lambda = 1$.

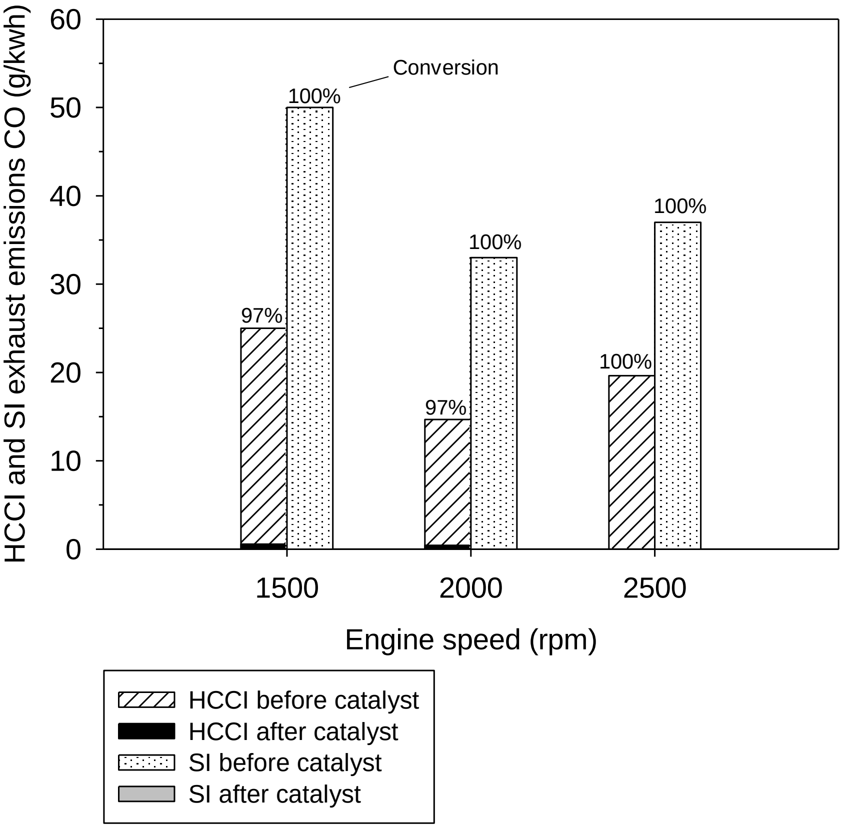

Figure 10: Catalytic conversion efficiency of CO, (4bar NMEP), HCCI, SI, λ = 1.

Table 1: Engine specifications.

Engine Specification	Details
Engine type	4 valve per cylinder, V6
Intake valve timing	In MOP 500 CA
Exhaust valve timing	Variable
Bore x Stroke	89.0 mm × 79.5 mm
Intake temperature	300 k
Compression ratio	11
Air / fuel ratio	14
Exhaust Throttled	15 mm diameter orifice
Valve lift HCCI mode (all valves)	3 mm

Table 2: Engine test conditions (low load).

Air/Fuel ratio (λ)	Engine Speed (rpm)	Exh. Temp. (°C)	Oxygen (%)	Load NMEP (bar)	GHSV (kh^{-1})
1.2	1500	370	0.02	3	25
	2000	355	0.01	3	28
	2500	340	0.01	3	23
1.4	1500	350	0.20	3.5	26
	2000	355	0.30	3.5	29
	2500	345	0.90	3.5	28
1.6	1500	355	1.00	4	31
	2000	350	1.50	4	33
	2500	360	1.40	4	34

Table 3: Engine test conditions (higher load).

Air/Fuel ratio (λ)	**Engine Speed (rpm)**	**Exh. Temp. (°C)**	**Oxygen (%)**	**Load NMEP (bar)**	**GHSV (kh^{-1})**
1.2	1500	400	0.02	4	27
	2000	405	0.01	4	29
	2500	420	0.04	4	31
1.4	1500	375	0.20	4.5	29
	2000	382	0.80	4.5	31
	2500	390	0.90	4.5	32
1.6	1500	370	1.40	5	32
	2000	393	1.50	5	35
	2500	385	1.30	5	26

Table 4: Engine conditions and exhaust emissions under HCCI and SI modes (λ=1) at different engine speeds.

Engine conditions			**SI Mode** **Exhaust emissions (g/kwh) before catalyst**						**HCCI Mode** **Exhaust emissions (g/kwh) after catalyst**					
Cond	**Speed rpm**	**load bar**	**HC CH_3 Equiv**	**CO**	**NO_x**	**Exh temp (°C)**	**O%**	**Fuel cons (g/hr)**	**HC CH_3 Equiv**	**CO**	**NO_x**	**Exh temp (°C)**	**O%**	**Fuel cons (g/hr)**
1	1500	4	1.80	50	4.60	590	0.80	3712	4.30	25	0.70	409	1.40	3484
2	2000	4	1.30	33	6.80	636	1.10	4926	1.90	14	1.60	422	0.95	4228
3	2500	4	1.10	37	10.0	670	1.10	5494	1.50	19	1.40	444	0.65	5128