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Development of an Ultra Compact CPOX Reactor for Diesel Fuel

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1 Background

Environmental conservation awareness is increasing all over the world. As a result, intensification of emission regulation in automotives has become more stringent, and it has been extended not only to HC, CO, NOx, and particulates, but also to CO_2. Therefore, further improvement of automotive exhaust gas emission and fuel consumption are required for internal combustion (IC) engines. This includes Diesel Engines, which have a high market share in EU because of its high torque and low fuel consumption. Even diesel engines will be required to achieve lower exhaust emissions and improved fuel economy. However, it is difficult to further reduce diesel engine emissions with existing technology. Hence, novel technical solutions are required.

One way to address these issues is by improving combustion and after-treatment efficiency by dosing hydrogen and/or other reducing-gas into the engine intake and/or upstream of the exhaust catalyst [1,2]. To do this an on-board reforming reactor, which can synthesis hydrogen and/or other reductant gas using diesel fuel, is needed.

New powertrains, such as Partial Hybrid Electric Vehicles (PHEV), are an alternative approach towards meeting these stringent emission regulations. Especially, Series HEVs, in which the engine is used only for power generation i.e. as an Auxiliary Power Unit (APU), is one of the most effective systems for reduction of CO_2 emissions. In addition, recent approaches which replace the IC engine in favour of fuel cells such as Solid Oxide Fuel Cells (SOFC) with onboard reforming further improve power generation efficiency [3] .

Reforming reactors however have been primarily developed for stationary applications. The key requirements for these reforming reactors include high conversion of fuels into hydrogen and durability at constant load for tens of thousands of hours. Generally, steam reforming (SR) and auto thermal reforming (ATR) are chosen for those uses [4,5]. These reaction products contain higher concentrations of hydrogen via endothermic reactions or heat-independent reactions from hydrocarbon fuels, steam, and air, as needed.

In addition to the above features, high adaptability to transients e.g. start up and turndown, low HC in syngas, compactness, reasonable cost, and sufficient durability despite transient operation must also be realized in consideration of aspects required for automobile use. There is also a difference in the fuels of interest and lack of water. For instance, methane or natural gas is the primary fuel used for stationary applications, but for on-board applications, a liquid fuel such as diesel or gasoline has to be used in keeping with the existing fuelling infrastructure.

A diesel-fuelled, experimental reforming reactor, suitable for on-board automobile applications, was developed. The goal was to design a reforming reactor that was distinctly different from conventional reformers for stationary or fuel cell applications.

2 Reforming Reactor

In order to satisfy the requirements for such an on-board reformer, a reforming reactor was designed and fabricated. It had two novel features. The first was waterless CPOX of diesel on a radial configuration. The second was a suitable fuelling strategy capable of operating under rapid transients.

2.1 Reactor design

The key design emphasis was on compactness and high conversion of fuel to hydrogen. Dry or waterless CPOX, which produces hydrogen using only fuel and air was chosen as the reforming approach. This permits use of a simple reforming reactor by avoiding use of steam, while performing at a relatively high reforming efficiency because of a use of catalyst.

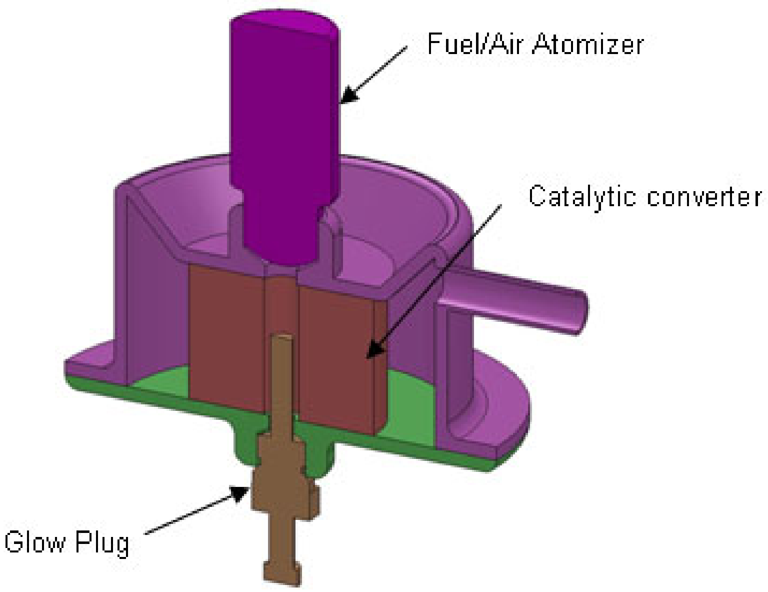


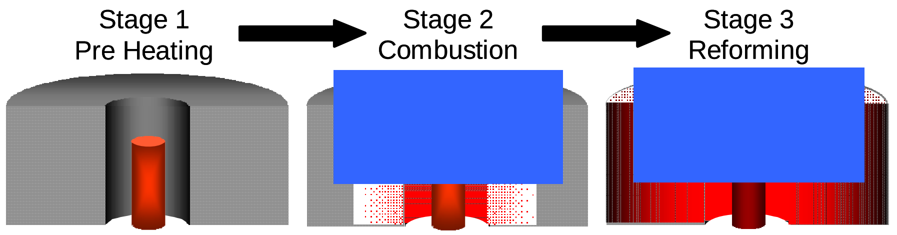
Figure 1: Schematic diagram of a radial flow dry CPOX reforming reactor.

Figure 1 shows a schematic diagram of the CPOX reforming reactor. This reactor comprises primarily of a catalytic converter which produces hydrogen using the fuel and air, a glow plug capable of raising the temperature of the catalytic converter to activation temperature quickly, and an atomizer capable of uniformly mixing fuel and air. These components are additionally arranged within close proximity. Consequently, the reactor was extremely compact at a total volume of approximately 150 cc (catalyst size: 20 cc). In order to achieve compactness, a radial flow design was adopted in which the flow of inlet gas to the catalytic converter passes radially through the catalyst. In order to realize a radial flow system, a substrate with a three-dimensional network structure was used [6,7]. There have been various reports on research into dry CPOX using diesel fuel, but those systems generally had a heating section to heat up the air before introducing it to the reactor, or a vaporizing section and mixing section to make a uniform mixture of fuel and air to achieve fast startability and high conversion to hydrogen. The developed reactor, by contrast, is able to achieve these properties required without these sections by using an atomizer to enable a very fine and uniform mixture of fuel and air in a compact space.

2.2 Fuelling strategy in transient state

Transient reformer operation, such as start up and syngas flow rate change, requires rapid and stable operation with minimum higher HC breakthrough. We employed a fuelling strategy to enable this. It is represented in Figure 2. The operation consists of three stages and is dependent on the temperature of the catalytic converter.

In Stage 1 (pre heating), the glow plug is switched on and heats the catalytic converter until it reaches the catalyst activation temperature (in the figure, it is posited as 400 °C). At this time, neither fuel nor air has been fed into the reactor. Next, in Stage 2 (combustion), fuel and air are co-fed. Here, the catalyst is further heated by the combustion reaction. In this step, fuel and air flow rate and O/C ratio are adjusted for lean operation for rapid heat up with low HC emission. This permits quick transition to Stage 3 (reforming). Finally, when the temperature of the catalytic converter reaches the optimal temperature (in the figure, it is posited as 800 °C) suitable for dry CPOX reaction, operation shifts to Stage 3 and the fuel and air flow rate is changed to a condition in which the dry CPOX reaction is efficiently sustained by heat-independent operation. The power to the glow plug is also switched off and the system shifts to steady-state. In Stage 3, the dry CPOX reaction proceeds sufficiently and the temperature range is set (this time from 800 °C to 1,100 °C) so as to avoid thermal degradation of the catalyst. Syngas flow rate change is implemented by varying the fuel flow rate while keeping within this temperature range. Despite the aforementioned strategy, the duration of Stage 2 during transient operation was a few seconds. Further reduction in response time was made possible via the use a quick response solenoid valve as the fuel flow controller.



Temp. on the upper of the catalyst	RT～400℃	400℃～800℃	800℃~1100℃
Glow plug	ON	ON	OFF
Fuel/air	Not fed	fed	fed
O/C	–	Lean than STEP3	about 1.0

Figure 2: Image of the control logic on a dry CPOX reforming reactor.

3 Performance

3.1 Steady state performance

Figure 3 shows the steady state performance result at several fuel and air flow rates and O/C ratios. This result shows that an optimal flow rate of fuel and air exists in order to achieve high hydrogen production efficiency.

Conditions which showed best performance (i.e. high hydrogen concentration and low THC concentration) were at a fuel flow rate=3.81 g/min., air flow rate=17.0 L/min., O/C=1.05 (Figure 3) with hydrogen concentration of 20.9 %. This corresponds to greater than 80 % hydrogen yield and to about 5L/min. of hydrogen production. In addition, THC concentration at this condition was at 0.65 %.

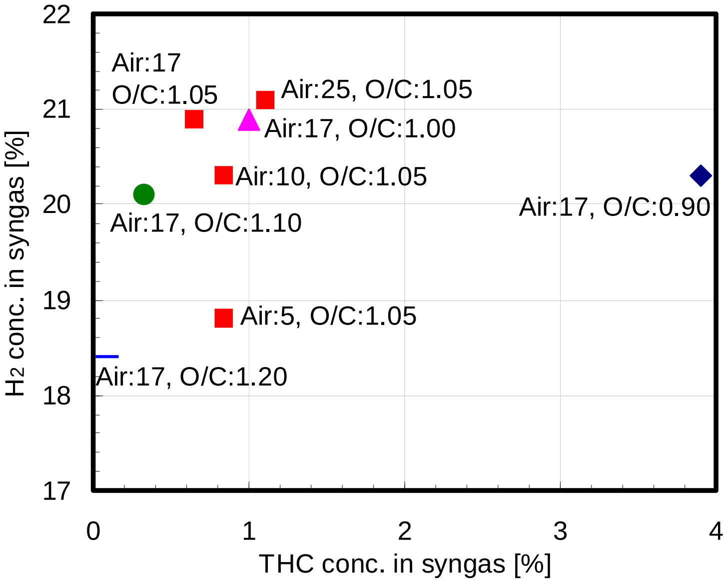


Figure 3: Steady state performance of dry CPOX at several fuel/air condition.

3.2 Transient performance

With the aforementioned strategy, start up performance was evaluated. Operating condition for each Stage is shown in Table 1. Note that the value of O/C in Stage 2 was higher than in Stage 3.

Table 1: Operation condition on transient performance evaluation.

	Stage 2	Stage 3
Fuel	1.77 g/min	3.81 g/min
Air	15 L/min	17 L/min
O/C	2.00	1.05
Operation range	400 ~800	800 ~1000

3.3 Start up performance

During start up, the temperature rises quickly and reaches reforming temperature without overshoot. Hydrogen concentration increase follows the temperature ramp. Start up time was defined as the time taken to reach 90 % of steady state hydrogen concentration. This period was observed to be 30 seconds after cold start at room temperature. Figure 4 shows the behaviour of hydrogen concentration in syngas and temperature on catalytic converter during start-up performance test.

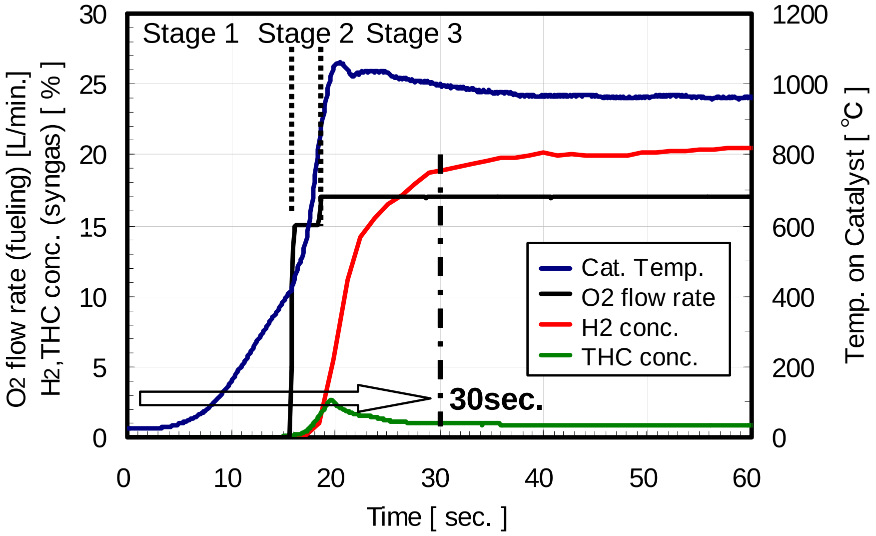


Figure 4: Start up performance of dry CPOX by our fuelling strategy.

3.4 H2 production change performance

Next, the rate of change of hydrogen production was examined as a function of the fuelling strategy used during Stage 3. The maximum and minimum hydrogen production rates were identified (Table 2).

Table 2: Operation condition and result on H_2 production change performance evaluation.

	Max H_2 production	Min H_2 production
Fuel	3.81 g/min	1.90 g/min
Air	17 L/min	8.5 L/min
O/C	1.05	1.05
Cat. Temp.	1000 (approx.)	950 (approx.)
H_2 concentration	20.9 %	19.8 %
H_2 production	1.05	1.05

Figure 5 shows the transient behaviour of hydrogen and THC concentration in syngas, fuelling air flow rate, and temperature within the catalytic converter. Hydrogen production changed quickly and in a stable manner form 2.3L/min. to 5.2L/min. No transient temperature spikes or other such phenomena were observed. This was not possible with slow response fuel controllers.

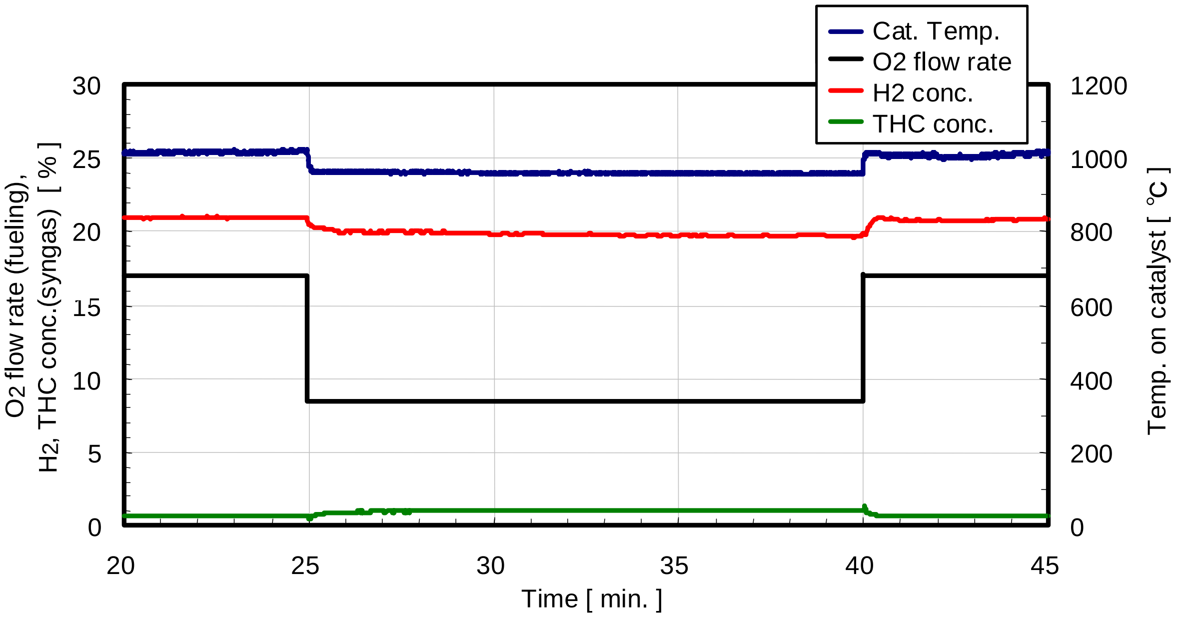


Figure 5: **H_2 production change performance behaviour of dry CPOX.**

4 Conclusion

Experimental development of an on-board diesel reforming reactor suitable for an automobile was completed. A reforming reactor was demonstrated with high conversion to hydrogen, good transient response, including start up and syngas flow-rate change, and low HC breakthrough within an ultra compact package. This work confirmed the possibility of on board reforming for automobile applications.

References

[1] L. McWilliam, T. Megaritis, H. Zhao, Experimental Investigation of the Effects of Combined Hydrogen and Diesel Combustion on the Emissions of a HSDI Diesel Engine, SAE Paper 2008-01-1787 (2008)

[2] K. Theinnoi, S. Sitshebo, A. Tsolakis, V. Houel, R. R. Rajaram, Hydrogen Promotion of Low-Temperature Passive Hydrocarbon-Selective Catalytic Reduction (SCR) over a Silver Catalyst, Energy Fuels Vol22 No.6 4109-4114 (2008)

[3] J. J. Botti, M. J. Grieve, J. A. MacBain, Electric Vehicle Range Extension Using an SOFC APU, SAE Paper 2005-01-1172 (2005)

[4] R. Gerd, H. Viktor, Hydrogen for fuel cells from ethanol by steam-reforming, partial-oxidation and combined auto-thermal reforming: A thermodynamic analysis, J. Power Sources Vol.185 No.2 1293-1304 (2008)

[5] J. Pasel, J. Meissner, Z. Pors, R.c. Samsun, A. Tschauder, R. Peters, Autothermal reforming of commercial Jet A-1 on a 5kWe scale, Int. J. Hydrogen Energy Vol. 32 No.18 4847-4858 (2007)

[6] S. Roychoudhury, M. Castaldi, M. Lyubovsky, R. LaPierre, S. Ahmed, Miclrolith catalytic reactor for reforming *iso*-octane-based fuels into hydrogen, J. Power Sources 152 75-86 (2005)

[7] M. Lyubovsky, H. Karim, P. Menacherry, S. Boorse, R. Lapierre, W. C. Pfefferle, S. Roychoudhury, Complete and partial catalytic oxidation of methane over substrates with enhanced transport properties, Catal. Today Vol.83 No.1/4 183-197 (2003)