Effects on CHP plant efficiency of H 2 production through partial oxydation of natural gas over two group VIII metal catalysts

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EFFECTS ON CHP PLANT EFFICIENCY OF H_2 PRODUCTION THROUGH PARTIAL OXYDATION OF NATURAL GAS OVER TWO GROUP VIII METAL CATALYSTS

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ABSTRACT

Blending H_2 with natural gas in spark ignition engines can increase for electric efficiency. In-situ H_2 production for spark ignition engines fuelled by natural gas has therefore been investigated recently, and reformed exhaust gas recirculation (RGR) has been identified a potentially advantageous approach: RGR uses the steam and O_2 contained in exhaust gases under lean combustion, for reforming natural gas and producing H_2, CO, and CO_2. In this paper, an alternative approach is introduced: air gas reforming circulation (AGRC). AGRC uses directly the O_2 contained in air, rendering the chemical pathway comparable to partial oxidation. Formulations based on palladium and platinum have been selected as potential catalysts. With AGRC, the concentrations of the constituents of the reformed gas are approximately 25% hydrogen, 10% carbon monoxide, 8% unconverted hydrocarbons and 55% nitrogen. Experimental results are presented for the electric efficiency and exhaust gas (CO and HC) composition of the overall system (SI engine equipped with AGRC). It is demonstrated that the electric efficiency can increase for specific ratios of air to natural gas over the catalyst. Although the electric efficiency gain with AGRC is modest at around 0.2%, AGRC can be cost effective because of its straightforward and inexpensive implementation. Misfiring and knock were both not observed in the tests reported here. Nevertheless, technical means of avoiding knock are described by adjusting the main flow of natural gas and the additional flow of AGRC.

Keywords: Hydrogen, CHP, natural gas, power,

1. INTRODUCTION

Efforts to improve the performance of internal combustion (IC) engines (e.g., brake mean effective pressure, specific fuel consumption) are nowadays limited by both stringent emissions regulations and abnormal combustion problems (knock, engine misfiring). These limitations are particularly important for lean burn natural gas engines that are used for combined heat and power (CHP) applications. Examples of such constraints are shown in Figure 1, where engine performance parameters are plotted against equivalence ratio. Improving the electric efficiency of stationary IC engines fuelled by natural gas should not cause any of the above noted constraints to be exceeded. The window between knock and combustion misfire is narrow, as shown in the left diagram of Figure 1, and can occur for an equivalence ratio in the range 1.7 to 2.0, common tuning for CHP applications. Nevertheless, specific emissions are relatively low in this equivalence ratio range, as shown in the right diagram of Figure 1, suggesting that a good compromise among these factors is attained by operating in this equivalence ratio range.

Fig. 1. Variation of selected engine performance parameters with equivalence ratio, highlighting challenges (firing problems, emissions) in improving engine performance

It has been demonstrated experimentally by Le Corre et al. [1] that the addition of H_2 has a positive effect on IC engines fuelled by traditional fuels. Increased H_2 content increases the mechanical efficiency of such IC

engines without major increases in engine emissions. The main hindrance related to this measure is the requirement for H_2 storage cylinders, especially for transport applications.

To comply with emissions regulation two main approaches exist according to Einewall et al. [2]: operating under stoichiometric conditions with exhaust gas recirculation and a three-way catalyst, or operating under lean burn conditions.

In-situ H_2 production by steam reforming has been considered for IC engines fuelled by natural gas (e.g., Yap et al. [3]). Such applications are suitable for stationary CHP plants which are not subject to significant weight and volume constraints. In-situ steam reforming involves the following:

1. The catalytic reforming device is located in the exhaust gases in order to keep it at high temperature (775 K).
2. Part of the exhaust gases is used to supply the catalyst with water vapour, oxygen and carbon dioxide,
3. Reformed gases (enriched in hydrogen) are then mixed with the intake air and gas line, as is done with exhaust gas recirculation (EGR) technology used in the car industry to reduce NO_X emissions for diesel engines.

Le Corre et al. [1] highlighted the effect of air-fuel ratio on the H_2 production for a CHP plant. The higher the air-fuel ratio, the greater is the H_2 content in the reformed gases. For example, Le Corre et al. [1] reported that the difference of H_2 content is 2% for air-fuel ratios of 1.5 and 1.4. This observation implies that the O_2 content limits reactions where it is a reactant. Two such reactions are the complete oxidation of methane and the partial oxidation of methane:

Complete oxidation of methane: $CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$ ($\Delta H_{298\,K} = -890\ \mathrm{kJ\ mol^{-1}}$) (1)

Partial oxidation of methane: $CH_4 + ½O_2 \rightarrow CO + 2H_2$ ($\Delta H_{298\,K} = -35.6\ \mathrm{kJ\ mol^{-1}}$) (2)

The individual reactions that contribute to the overall reaction consist of the reforming of CO_2 into CO (equation 3), the steam reforming of methane into CO (equation 4), the water gas shift reaction (equation 5), the oxidation of hydrogen (equation 6) and the oxidation of carbon monoxide (equation 7):

$CH_4 + CO_2 \rightarrow 2CO + 2H_2$ (3)

$CH_4 + H_2O \rightarrow CO + 3H_2$ (4)

$CO + H_2O \rightarrow CO_2 + H_2$ (5)

$2H_2 + O_2 \rightarrow 2H_2O$ (6)

$2CO + O_2 \rightarrow 2CO_2$ (7)

For a CHP plant fuelled by natural gas, the O_2 content in the exhaust gases is around 7-8% by vol. In this configuration for H_2 production, hydrogen makes up around 10-14% by vol. of dry reformed gases. Then, O_2 content should be an important reactant.

Note that the exhaust gases from natural gas CHP plant typically contain three times less than the O_2 concentration in the air. This circumstance raises a question: Could the system be modified in a way that allows the exhaust gases to be substituted for some of the additional air provided at the reformer inlet?

The partial oxidation of methane has many advantages compared to steam reforming of methane for hydrogen production. An exothermic reaction, the partial oxidation of methane produces syngas with a H_2/CO volumetric ratio of about two, which is ideal for further methanol and Fischer-Tropsch syntheses, according to Hadj-Sadok Ouaguenouni et al. [4]. Many precious metals, such as Ru, Rh and Pt, have been tested as catalysts for the partial oxidation of methane reaction (see Torniainen et al. [5]; Schmidt and Huff [6]). Schmidt et al. [7] and Deutschmann and Schmidt [8] have proposed a model to understand the complex interaction between transport and kinetics involved in the partial oxidation of methane. Some authors have tested a commercial nickel-based catalyst, but this approach was rejected due to activation difficulties. Pd and Pt have been identified as advantageous potential catalysts, and are considered here.

The main objective of this paper are to investigate the use of partial oxidation of methane as a means of improving H_2 production in CHP plant applications, and to examine how the process occurs in the presence of two group VIII metal catalysts (Pt and Pd). It is recognized that this approach must be cost effective for it to be adopted in real applications.

The concept considered here, which is referred to as air gas reforming circulation (AGRC), is shown in Figure 2. A catalytic reforming device is placed in the exhaust gas flow, but the catalysts do not come into contact with the exhaust gas, as it is used only to heat the catalyst. A chemically active blend of air and natural gas is located over the catalyst. The reformed gases are then mixed with the fresh mixture of fuel and air at the engine inlet. The results are reported in this paper of experimental studies in which two DCL© catalysts are examined: one based on Pt and one on Pd.

Legend: (10) air flow meter, (11) air filter, (12) carburetor, (13) turbocharger, (14) intercooler, (15) actuator, (16) engine, (17) catalyst device, (18) exhaust heat exchanger, (20) natural gas flow meter, (21) Methane Number sensor, (30) additional air flow meter, (40) additional natural gas flow meter, (50) methane number sensor.

Fig. 2. Engine system incorporating the air gas reforming circulation (AGRC) concept.

2. EXPERIMENTAL APPARATUS AND PROCEDURE

The test bench is a spark ignition (SI) engine fuelled by natural gas (see Figure 3). This installation is a combined heat and power plant operating at a fixed engine speed, as specified in Table 1.

Table 1. Engine specifications.

Fig. 3. Reforming device installation (at left) on a 210 kWe CHP gas engine (orange).

The engine generates a constant electrical power of 210 kW during operation. This power output is kept constant by a control loop that acts on a butterfly valve which adjusts the air-gas flow entering the cylinders. The air-fuel ratio is controlled manually by acting on the output of pressure reducer upstream of the carburetor using the Venturi effect [9]. The engine spark advance is kept constant throughout the testing, at 14 CA BTDC.

The test bench is equipped with various experimental sensors, corresponding to low frequency measurements (1 Hz). Measurement ranges are shown in parentheses.

- Thermocouples to measure the catalyst inlet and outlet temperature, and exhaust gas inlet and outlet temperature in the catalyst housing.
- A mass flow rate meter for the main natural gas SI engine feed line (0-1200 Nm^3/h), and a mass flow rate meter for the additional natural gas at the catalyst inlet (0-600 Nm^3/h).
- A mass flow rate meter for the intake air (0-1500 kg/h).
- Two dry-basis gas analysers:
  - Engine exhaust gases are analysed with the following:
    - HORIBA VA300 analyser: O_2 (0-25%), CO_2 (0-20%) CO (0-1%), NO (0-2000 ppm)
    - COSMA Cristal 500 analyser: HC (0-5000 ppm)
  - Catalyst outlet gases are analysed with the following:
    - ROSEMOUNT NGA2000: H_2 (0-30%), O_2 (0-30%), CH_4 (0-30%), CO (0-30%), CO_2 (0-30%)
    - SIEMENS H3-600: CO_2 (0-16%)
- A wattmeter (ENERNIUM-CFG1).
- A mass flow rate meter for the water cooling flow rate of SI engine (0-50 m^3/h).
- PT sensors to measure the temperatures at several points in water cooling circuit of the engine (PT100, 0-200℃).

All data were averaged over a period of 15 minutes once steady state operation was achieved. The H_2-enhanced engine electrical efficiency η_e is defined as follows:

$$\eta_e = \frac{W_e}{LHV\,(q_{NG} + q_{NGR})} \tag{8}$$

Here, q_{NG} denotes the natural gas flow rate (main stream), q_{NGR} the natural gas flow rate for reforming, and LHV the lower heating value of natural gas. The two natural gas flows are shown in Figure 2.

The catalytic reforming device is illustrated in Figure 4.

The following experimental procedure was used for the catalytic reforming device shown in Figure 4:

1. The additional air flow rate over the catalytic reforming device was set.
2. The additional natural gas flow rate at catalyst inlet was set. Consequently, part of natural gas is not converted; this is not a disadvantage of the system since the main natural gas flow rate is decreased correspondingly.
3. The equivalence ratio of the SI engine is kept constant by modifying the air flow rate in the main stream. Note that the O_2 content in the exhaust line is measured, and provides a straightforward means of maintaining the same condition during the combustion phase.

Fig. 4. Schematic of catalytic device. UHC denotes unburned hydrocarbons.

Two commercial honey-comb catalysts of DCL© are tested. These are made of formulations based on Pd and Pt, which are proprietary information of DCL©. Selected features of the catalysts are listed in Table 2.

Table 2. Features of tested catalysts

For safety reasons, the following measures were adopted:

- The system is operated at 80% of its nominal electric power output. This engine operation is less demanding than operation with a full fuel charge.
- Some part of additional natural gas flow rate over catalyst does not react. That is, oxygen is completely consumed at the catalyst outlet. This operational condition for the catalyst is safer since it permits auto-ignition of H_2 to be avoided.

The volumetric flow rate over the honey-comb catalyst is a blend of additional air and natural gas for reforming, denoted NGR. Two flow rates for “additional air” are used: 3 and 5 Nm^3/h. The range of the volumetric ratio NGR/O_2 is bounded between 0.3 and 1.6, regardless of the catalyst, as shown in Figure 5. This volumetric ratio represents the ratio of the volumetric flow rate of natural gas for reforming to the volumetric flow rate of oxygen, in the overall gas flow. It is noted that, in the literature on steam reforming of natural gas, results are typically based on the ratio NGR/H_2O (e.g., Peucheret et al. [10]). Nonetheless, in this paper, the significant ratio is NGR/O_2.

Fig. 5. Experimental design of flow rates of additional air over the catalyst in the feed line of the SI engine, for various NGR/O_2 ratios and catalysts.

3. RESULTS AND DISCUSSION

3.1 Catalyst performance in AGRC

The first results presented concern exclusively the catalyst performance when Air Gas Reforming Circulation (AGRC) is used. All results are given on dry basis. Natural gas and air react chemically over the catalyst; N_2 is the constituent with the highest concentration at the catalyst input and output. We provide results for H_2, CO and unconverted natural gas. To avoid H_2 auto-ignition, as explained previously, the experiment is designed to ensure there is no O_2 at the catalyst output. The CO_2 concentration is around of 6-7% at the catalyst output.

The primary gases at the exit of the catalyst are shown in Figure 6. In the top two graphs of Figure 6, H_2 production is seen to depend on the catalyst formulation, and to result in hydrogen concentrations of approximately 26% for the Pd catalyst and 21% for Pt. H_2 production also depends on the air flow rate, indicating that fluid dynamic effects are very significant. In the configuration with reformed exhaust gas recirculation (denoted RGR), where exhaust gas is blended with additional natural gas before entering the reformer, Le Corre et al. [1] have shown that H_2 production yields a concentration in the reformed gas of only 10-14%. The AGRC improves significantly H_2 production by promoting the partial oxidation of methane, at temperatures around 775 K. This temperature is not the most favorable for H_2 production (the ideal temperature for H_2 production is around 1000 K); but the exhaust gases exiting the turbocharger are available at 775 K for use and are otherwise emitted as wastes.

In Figure 6 (middle two graphs), CO production is observed to be significant, leading to a CO concentration of around 11% for the Pd catalyst and 8% for Pt. Such amounts are not a major problem in IC engines, contrary to fuel cell applications. As for H_2 production, CO production also depends on the air flow rate, indicating that fluid dynamic effects are very significant, as is the chemical pathway, denoted by equations 1-6, over the catalyst.

In Figure 6 (bottom two graphs), the concentration of unconverted natural gas content is shown, demonstrating that O_2 (from additional air) is totally consumed, avoiding the risk of H_2 auto-ignition. When the additional air flow rate is 3 Nm^3/h, the experimental facilities are not capable of measuring a NGR/O_2 ratio lower than 0.3. Clearly, however, the unconverted natural gas concentration is more important at an additional air flow rate of 3 Nm^3/h than 5 Nm^3/h.

Figure 7 illustrates the variation in H_2 concentration with CO concentration for the two catalysts and two air flow rates, and shows the role of catalyst composition (Pd and Pt). In Figure 5, ratios of reformed exhaust gas recirculation (NGR) to O_2 ranging between 0.3 and 1.6 are considered for the two air flow rates and the two catalysts. The concentrations of H_2 and CO, respectively, depend on air flow rate, achieving values over the Pd catalyst of about 26% and 11% at an air flow rate of 3 Nm^3/h and 20% and 8% at an air flow rate of 5 Nm^3/h. But H_2 and CO concentrations are not too sensitive to the ratio NGR/O_2 for an air flow rate of 3 Nm^3/h. Conversely, over the Pt and Pd catalysts, the H_2 and CO concentrations increase approximately linearly with the NGR/O_2 ratio. This observation suggests that chemical pathway is dependent on the catalyst choice.

Fig. 6. Variations in concentrations of main gases with the ratio NGR/O_2 of at the exhaust of catalyst for two catalysts and air flow rates.

Fig. 7. Variation of H_2 concentration with CO concentration for two catalysts and air flow rates.

3.2 Effects of catalysts on CHP system

The basic system (the CHP plant) has two inputs: natural gas and air in the main stream. The modified system (the CHP plant equipped with a catalyst device) is similar, except that the one natural gas stream is separated in two lines: one to the engine and the other to the catalyst. The absolute difference in electric efficiency is defined as the difference between the electric efficiencies of the modified and basic systems, with a positive absolute difference meaning that the catalyst device increases the electric efficiency and a negative value meaning that the catalyst device decreases the electric efficiency.

In Figure 8, the variation in the absolute difference of CHP electric efficiency with the ratio NGR/O_2 is shown for both Pd and Pt catalysts and for two air flow rates. In the left graph of Figure 8 for which the additional air flow rate is 3 Nm^3/h, the absolute difference is positive when the NGR/O_2 ratio is less than 1.3. In that figure, values for Pd (dotted spline line) and Pt (solid spline line) catalysts are seen to improve electric efficiency by similar amounts. In the left graph of Figure 8 for which the additional air flow rate is 5 Nm^3/h, the absolute difference of electric efficiency is approximately zero for low values of the ratio NGR/O_2 (< 0.8) and negative for high values of that ratio (> 0.8). Note that the results in Figure 8 and the related observations correspond with H_2 production concentrations of 26% at 3 Nm^3/h and of 20% at 5 Nm^3/h.

The benefit in electrical efficiency is directly attributable to the ability of H_2 to promote the combustion process. Several benefits of hydrogen combustion are described by Bauer and Forest [11]:

- The laminar flame speed for a stoichiometric hydrogen/air mixture (2.65-3.25 m/s) is about seven times higher than for methane or gasoline in air. This property of hydrogen leads to decreases the wall heat transfer to 17-25% of the primary fuel energy for hydrogen, compared to 22-33% for natural gas or 30-42% for gasoline.
- The “quenching distance” defined as the distance from the cylinder wall at which the flame quenches due to heat losses, characterizes the flame quenching property of a fuel in internal combustion engines. The quenching distance of hydrogen (0.064 cm at standard conditions) is approximately three times lower than that of other fuels, such as gasoline (0.2 cm at standard conditions) or methane (0.203 cm at standard conditions).
- Emissions from hydrogen fuelled engines are neither toxic nor photochemically reactive.

Fig. 8. Absolute difference of electric efficiency.

The uncertainty in the determined electric efficiency for the basic system (when there is no flow over the catalyst) is obtained by noting that the electric power W_e, the low heating value LHV and the mass flow rate of natural gas q_{NG} are three independent measures (measured by a wattmeter, a gas chromatograph and a mass flow meter, respectively):

$$d\eta_e = \frac{1}{LHV\ q_{NG}} dW_e - \frac{W_e}{LHV^2\ q_{NG}} d\ LHV - \frac{W_e}{LHV\ q_{NG}^2} d\ q_{NG} \tag{9}$$

The quadratic uncertainty $u(\eta_e)$ is defined based on the uncertainties of these three variables with its norm:

$$u(\eta_e)^2 = \left(\frac{1}{LHV\ q_{NG}} u(W_e)\right)^2 + \left(\frac{W_e}{LHV^2\ q_{NG}} u(LHV)\right)^2 + \left(\frac{W_e}{LHV\ q_{NG}^2} u(q_{NG})\right)^2 \tag{10}$$

$$\left(\frac{u(\eta_e)}{\eta_e}\right)^2 = \left(\frac{u(W_e)}{W_e}\right)^2 + \left(\frac{u(LHV)}{LHV}\right)^2 + \left(\frac{u(q_{NG})}{q_{NG}}\right)^2 \tag{11}$$

This is the classical law of uncertainty propagation see [12]. Substituting numerical values shows that the relative uncertainty of efficiency $\frac{u(\eta_e)}{\eta_e}$ is about 2.5%, with $u(W_e)/W_e = 2\%$, $u(LHV)/LHV = 1\%$ and $u(q_{NG})/q_{NG} = 1\%$. So the value of the electric efficiency η_e is 35% ± 1.0%.

In this investigation, the main instrumentation is shared between the basic system and the modified system (wattmeter, gas chromatograph for natural gas, and mass flow meter for natural gas in the main stream). The only difference is a second mass flow meter for the additional natural gas entering the catalyst. The uncertainty between these two configurations is evaluated, denoting the electric efficiency η_e^0 in the basic configuration and η_e^{NGR} when the catalyst device is active. The difference $\left(\eta_e^{NGR} - \eta_e^0\right)$ has been measured to be about 0.2%, which is equivalent to stating that the ratio $\frac{\eta_e^0}{\eta_e^{NGR}} \approx 0.99 < 1$. Here,

$$\frac{\eta_e^0}{\eta_e^{NGR}} = \frac{\frac{W_e}{LHV\ q_{NG_0}}}{\frac{W_e}{LHV\left(q_{NG_1} + q_{NGR}\right)}} \tag{12}$$

Where q_{NG_0} is the mass flow rate of natural gas in the basic configuration required to produce the electric power W_e, and q_{NG_1} is the mass flow rate of natural gas in the main stream of the modified configuration. Rearranging this equation yields

$$\frac{\eta_e^0}{\eta_e^{NGR}} = \frac{q_{NG_1} + q_{NGR}}{q_{NG_0}} \tag{13}$$

where q_{NG_0} and q_{NG_1} are measured by the same mass flow meter. As previously, the ratio $\frac{\eta_e^0}{\eta_e^{NGR}}$ is obtained by two independent measurements:

$$d\left(\frac{\eta_e^0}{\eta_e^{NGR}}\right) = \frac{q_{NG_0} dq_{NG1} - q_{NG_1} dq_{NG0}}{q_{NG_0}^2} + \frac{q_{NG_0} dq_{NGR} - q_{NGR} dq_{NG0}}{q_{NG_0}^2} \tag{14}$$

The fact that only one gas flow meter is used in the main stream implies that $dq_{NG_0} = dq_{NG1} \overset{not}{=} dq_{NG}$. Thus,

$$d\left(\frac{\eta_e^0}{\eta_e^{NGR}}\right) = \frac{q_{NG_0} - q_{NG_1}}{q_{NG_0}^2} dq_{NG} + \frac{q_{NG_0} dq_{NGR} - q_{NGR} dq_{NG}}{q_{NG_0}^2} \tag{15}$$

and its uncertainty is given by its norm:

$$u\left(\frac{\eta_e^0}{\eta_e^{NGR}}\right)^2 = \left(\frac{q_{NG_0} - q_{NG_1} - q_{NGR}}{q_{NG_0}^2} u(q_{NG})\right)^2 + \left(\frac{1}{q_{NG_0}} u(q_{NGR})\right)^2 \tag{16}$$

Substituting numerical values gives $u\left(\frac{\eta_e^0}{\eta_e^{NGR}}\right)$=5.04E-4. That means that the difference $\left(\eta_e^{NGR} - \eta_e^0\right)$ has an uncertainty of about 2.0E-4 and this difference is 0.2% ± 2.0E-4.

A time recording of electric efficiency and H_2 content is plotted on Figure 9 for the following conditions: spark timing advance 14CA BTDC, 7% O_2 content in the exhaust gases, 80% full load, ambient temperature 21 °C and relative humidity around 21%. Two gaps exist around 100 s and 1450 s. The first corresponds to turning off the additional natural gas flow, and the second one to turning it on. It is clear that uncertainties in the wattmeter or the gas chromatograph are not notable. Only the uncertainties of the two mass flow meters are important, as seen in equation 15.

Fig. 9. Time recording of electric efficiency and H_2 production.

An increase of electric efficiency is beneficial, but may not be acceptable if it increases exhaust gases emissions. In Figure 10, the variation in CO and HC emission concentrations in exhaust line of the CHP plant with the ratio NGR/O_2 are shown for the Pd catalyst and two air flow rates. It can be seen in the figure that the effect of the catalyst device is not significant on CO and HC emission concentrations, regardless of the additional air flow rate and the ratio NGR/O_2. Thus the increase of electric efficiency with the catalyst device does not appear to come at the expense of increased emissions, so a retrofit to achieve the absolute difference of electric efficiency is worth considering.

Fig. 10. Variation with NGR/O_2 ratio of the exhaust emission concentrations of the CHP plant equipped with a Pd catalyst device, for two air flow rates.

3.3 Discussion

Several processes used to improve the performance of engines are compared in Table 3. In this table, an SI engine fuelled by natural gas under lean conditions (taken to be an equivalence ratio of 1.4) equipped with a two-way catalyst (CO and unburned hydrocarbon UHCs oxidations) in the exhaust line is referred to as the Base case. Exhaust emissions of UHC, CO are considered after the two-way catalyst in the exhaust line. The modified engine cases considered include the Base case with exhaust gas recirculation (EGR), the Base case with reformed gas recirculation (RGR), and the Base case with air gas reforming circulation (AGRC). It is seen that the use of EGR and RGR processes improves engine environmental performance (especially NO_X emissions) but lowers energetic performance in terms of efficiency. However, the application of the AGRC concept to the GUASCOR engine increases the energy efficiency of the engine by around 0.2-0.4%, with little change in environmental performance. The results in Table 3 need to be validated on other types of engines having different control loops.

Table 3. Comparison of impact on technical and environmental performance of various engine modifications relative to a base case engine.

3.4 Engine knock issues and resolutions

The main risk of adding H_2 in internal combustion engines is the occurrence of knock associated with abnormal combustion. IC engines used for CHP installations usually run under strict operating conditions, usually based on achieving maximum electricity output while maintaining emissions at acceptable levels, as outlined in the Introduction. Hydrogen addition to natural gas decreases its relative methane content and is known to increase its ability to detonate. Hence, the knock tendency of an engine must be closely monitored when adding H_2 to increase engine efficiency.

Knock, which has been a concern since the invention of the IC engine, is caused by a local auto-ignition of gases under specific thermodynamic conditions, can seriously damage an engine (see Figure 11). Consequently, design and operating conditions are often limited by knock conditions.

Fig. 11. Piston damage from engine knock.

For knock problems, the methane number *MN* is commonly used to represent the gas quality, i.e. its ability to resist auto-ignition. It is usually equal to 100 for pure methane and 0 for pure hydrogen. This indicator is the equivalent of the Research Octane Number (*RON*) used for liquid fuels such as gasoline.

SI engines used as CHP plant can produce more than 1 MWe. Then, it is better to conceive a preventive protection instead of a curative one, as described by Le Corre et al. [13] and Saikaly et al. [14-16]. This is

especially true for applications involving hydrogen, where it is more advantageous to avoid knock rather than to detect it.

Knock conditions can be avoided by using AGRC, not to maximize the electric production but to protect the engine. Note that the methane number of reformed gas is calculated on the same basis as for natural gas, i.e., without inert gases. Since natural gas and reformed gas have two different methane numbers, it is possible to adjust the setting for the CHP plant to avoid knock conditions. This approach can form the basis of a preventive control mechanism.

A recent patent (number WO 2011010069) by Rahmouni and Le Corre [17] aims to avoid the occurrence of knock by using two methane number sensors (labeled 21 and 50 in Figure 1) to control the natural gas flow rates (both the main stream and the additional stream) to maintain an acceptable value entering the SI engine. These sensors are described in detail by Rahmouni et al. [18-20] and Loubar et al. [21].

Table 4. Methane number of natural gas and reformed gas

4. CONCLUSIONS

In this paper, air gas reforming circulation (AGRC) is proposed as a solution for in situ H_2 production for spark ignition engines fuelled by natural gas. Several important conclusions can be drawn from the results:

- With AGRC, the concentrations of the constituents of the reformed gas are approximately 25% H_2, 10% CO, 8% unburned hydrocarbons and 55% N_2. The AGRC reformed gas is blended with the main flow of natural gas and air, so unconverted natural gas is mixed with natural gas and is not problematic. The low heating value of CO is counterbalanced by the effect of H_2 during combustion in the cylinder of SI engine.
- The overall electric efficiency of an engine increases by 0.2% when the AGRC system is applied. Although the increase is not large, it is balanced by the fact that the AGRC system is simple to setup and cost effective.
- Equipping an SI engine with AGRC does not change significantly exhaust emissions (CO and HC) in comparison with the original configuration.
- AGRC appears to be advantageous compared to reformed gas recirculation because RGR decreases the electric efficiency of the overall system compared to an SI engine without reforming, while AGRC improves the electric efficiency.
- No knock or misfiring occurred during experimental tests. But, if abnormal conditions appear for some operating conditions, it is possible to adjust the natural gas main flow and the AGRC flow to avoid the problematic conditions by measuring and controlling the relative methane content of the combustion mixture so that it remains in the correct operating window, as defined by the engine manufacturer.

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List of figures

List of tables

Table 1

Table 1. Engine specifications.

Parameter	Value/Description
Engine manufacturer	GUASCOR FGLD 180
Number of cylinders	6
Bore	152 mm
Stroke	165 mm
Displaced volume by cylinder / total	2,994 / 17,964 cm^3
Clearance volume	300 cm^3
Compression ratio	11:1
Number of suction valves/exhaust valves	2 / 2 per cylinder
Valve train*: IO/IC/EO/EC	25 CA ATDC/ 45 CA ABDC/ 60 CA BBDC/ 15 CA BTDC
Turbo-charger pressure	1.8 bar
Engine speed	1,500 RPM
Ignition timing	14 CA BTDC

* CA: crankshaft angle; ATDC: after top dead center; BBDC: before bottom dead center; ABDC: after bottom dead center; BTDC: before top dead center; IO: inlet opens before TDC; IC: inlet closes after BDC; EO: exhaust opens before BDC; EC: exhaust closes after TDC.

Table 2

Table 2. Features of tested catalysts

Parameter	Value
Catalyst reference code	DC10LQ-1W10-21
Hourly space velocity (HSV)	28,000 h^{-1}
Length of honey-comb catalyst	9 cm
Diameter of honey-comb catalyst	21.7 cm

Table 3

Table 3. Comparison of impact on technical and environmental performance of various engine modifications relative to a base case engine.

Performance parameter	Modified engine cases		
	Base case with EGR	Base case with RGR	Base case with AGRC
Electrical efficiency	Decrease by 2% absolute	Decrease by about 1.5% absolute	Increase by 0.2-0.4% absolute
NOx emissions	Decrease	Same effect as EGR	No important effect
CO emissions	Increase	Same effect as EGR	No important effect
UHC emissions	Increase	Same effect as EGR	Slight decrease
Valve fouling	EGR valve fouling problem	RGR valve fouling problem	No fouling problem (no valve on exhaust line; standard valve on air line)

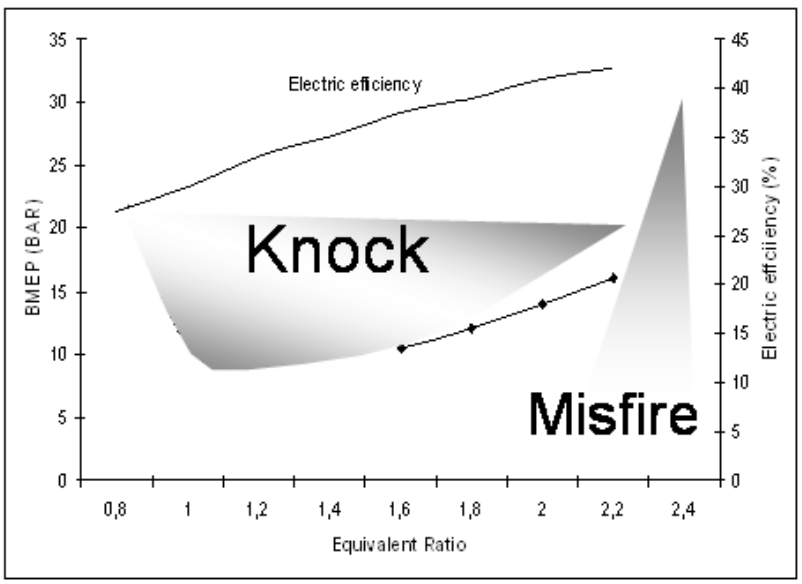
Table 4

Table 4. Methane number of natural gas and reformed gas

Content (vol. %)	Natural gas	Reformed gas with AGRC
O_2	0	0
CO_2	0.54	6.28
N_2	2.85	56.25
H_2	0	21.17
H_2S	0	0
CO	0	8.14
CH_4	87.68	8.16
C_2H_6	3.04	0
C_3H_8	5.6	0
C_4H_{10}	0.29	0
MN*	71.8	78.8

* Methane number is calculated based on the definition of Leiker et al. [22].

Figure 1


Electric efficiency
Knock
Misfire
BMEP (BAR)
Electric efficiency (%)
Equivalent Ratio
35
30
25
20
15
10
5
0
45
40
35
30
25
20
15
10
5
0
0,8
1
1,2
1,4
1,6
1,8
2
2,2
2,4

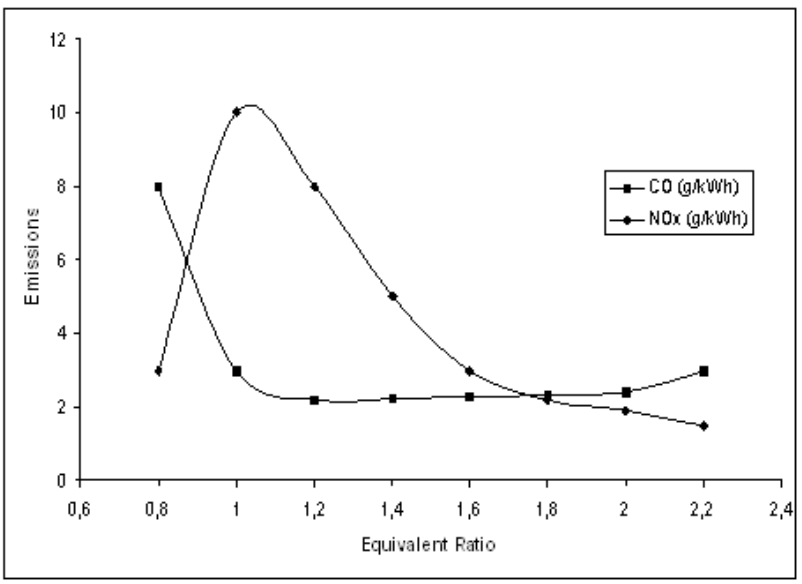

CO (g/kWh)
NOx (g/kWh)
Emissions
Equivalent Ratio
12
10
8
6
4
2
0
0,6
0,8
1
1,2
1,4
1,6
1,8
2
2,2
2,4

Figure 2

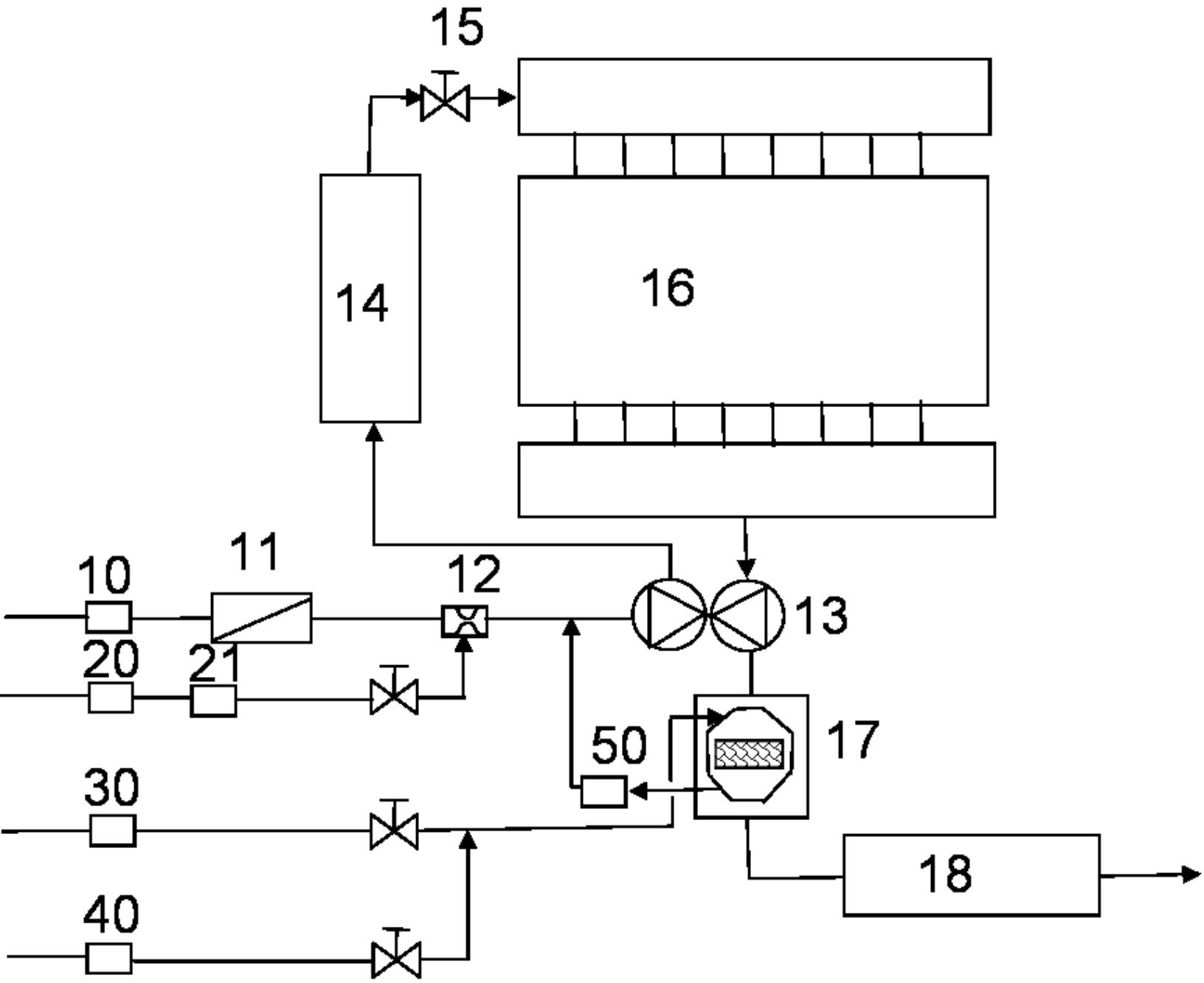


Figure 3


Natural gas
Main stream
Addtional
natural gas
towards catalyst device

Figure 4

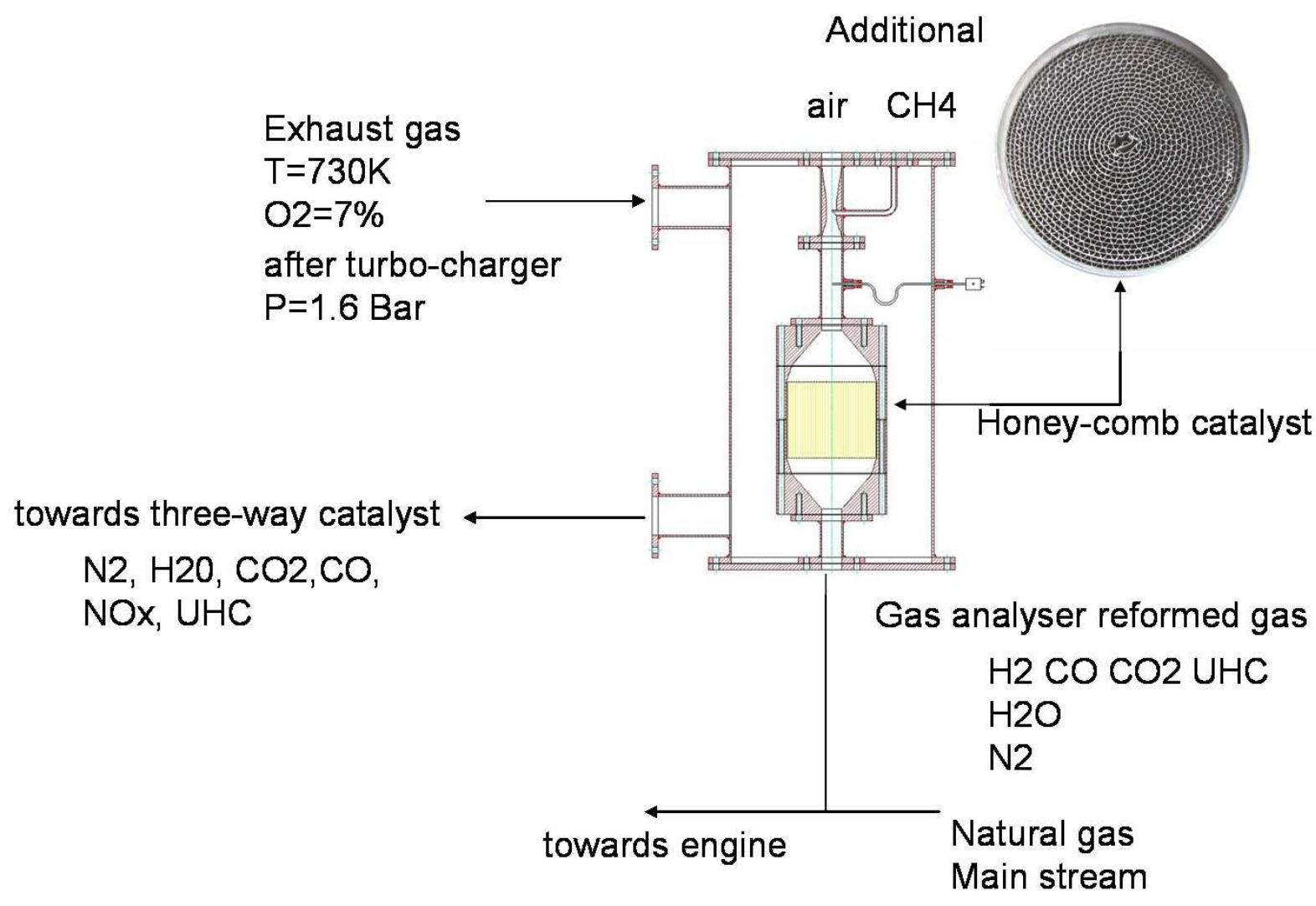

Additional
air CH4
Exhaust gas
T=730K
O2=7%
after turbo-charger
P=1.6 Bar
Honey-comb catalyst
towards three-way catalyst
N2, H20, CO2,CO,
NOx, UHC
Gas analyser reformed gas
H2 CO CO2 UHC
H2O
N2
towards engine
Natural gas
Main stream

Figure 5

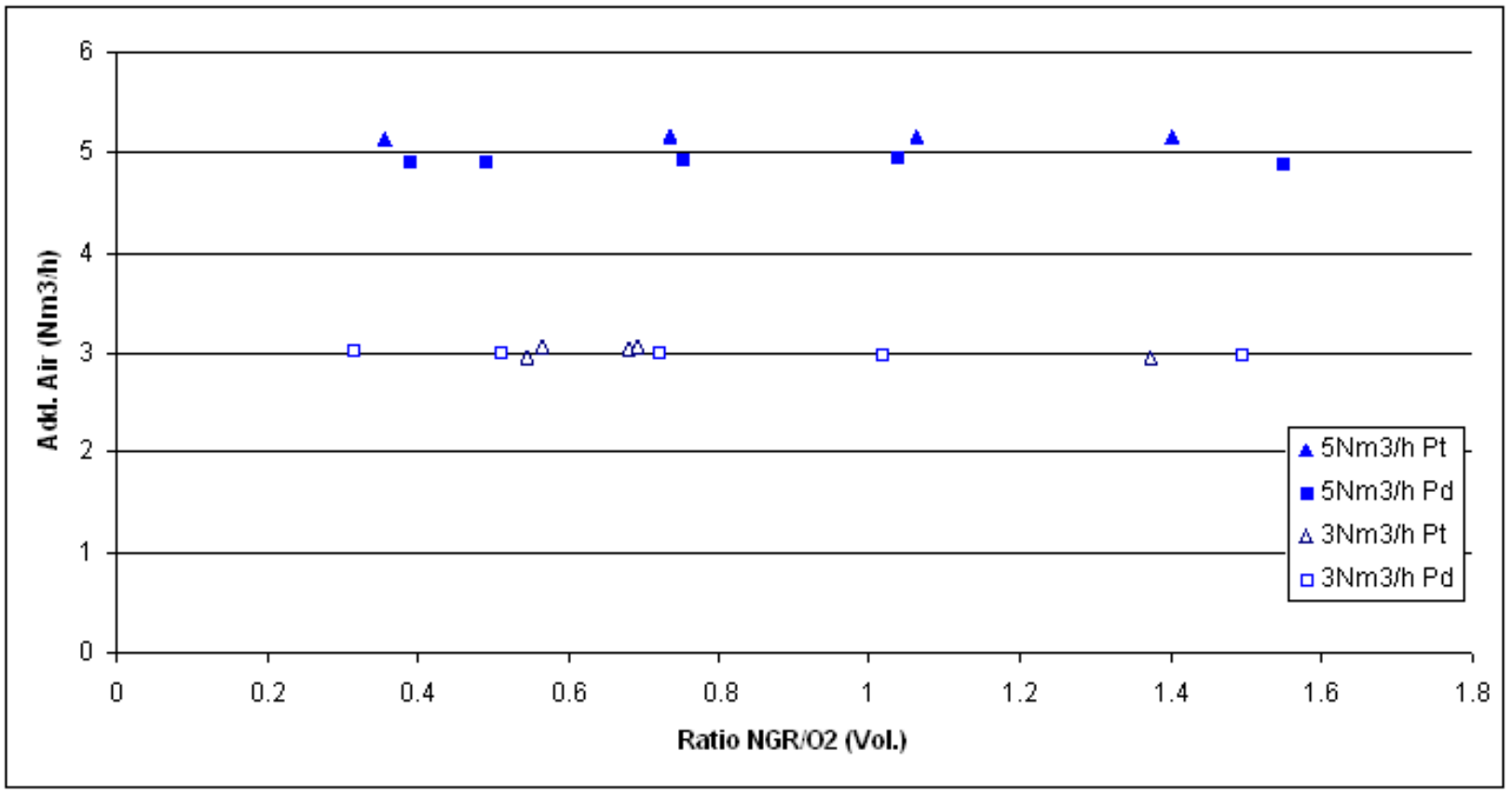
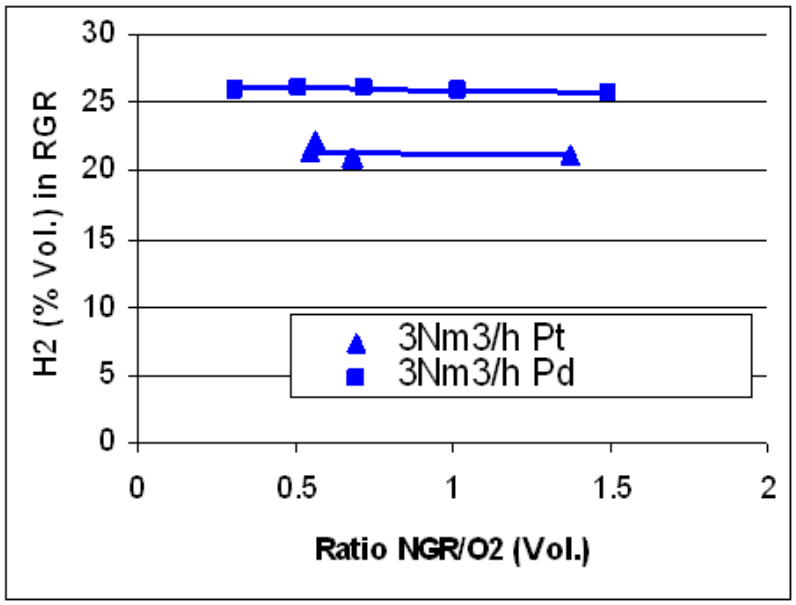
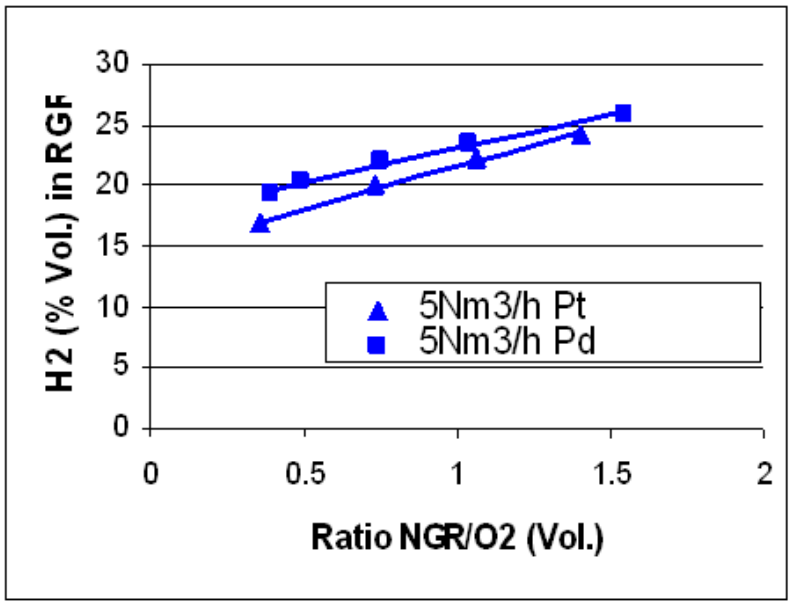
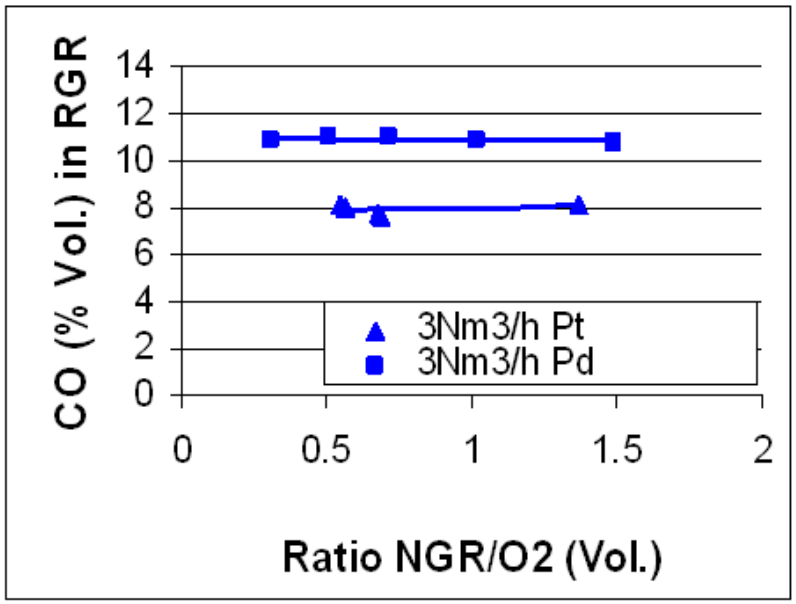
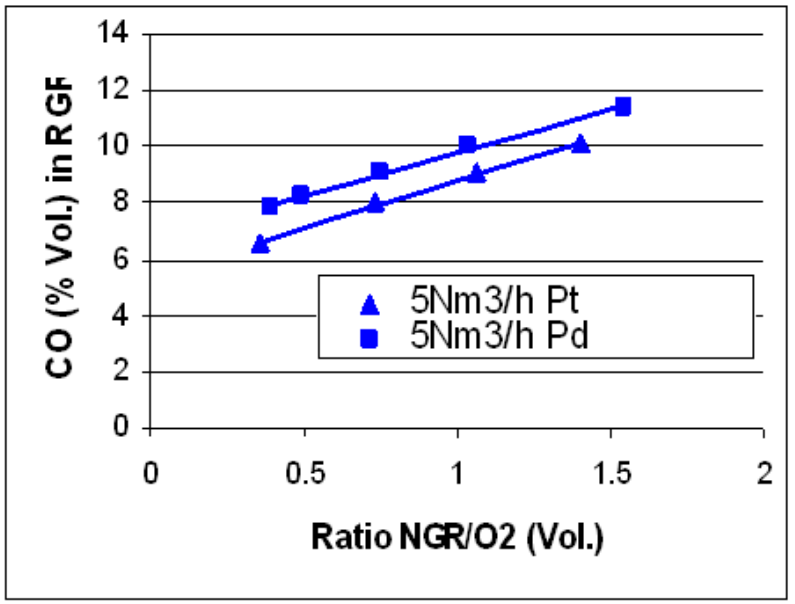

Add. Air (Nm3/h)
Ratio NGR/O2 (Vol.)
0
1
2
3
4
5
6
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
5Nm3/h Pt
5Nm3/h Pd
3Nm3/h Pt
3Nm3/h Pd

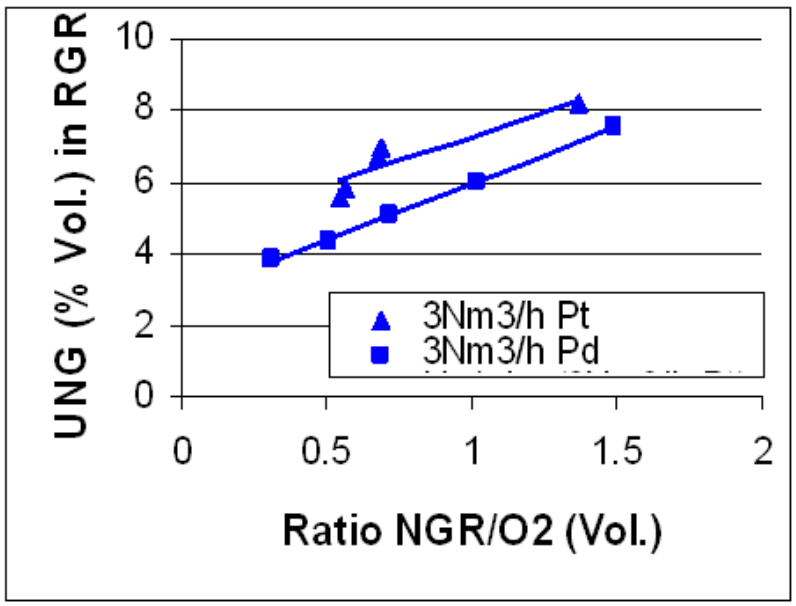
Figure 6


H2 (% Vol.) in RGR
30
25
20
15
10
5
0
0
0.5
1
1.5
2
Ratio NGR/O2 (Vol.)
3Nm3/h Pt
3Nm3/h Pd


H2 (% Vol.) in RGF
30
25
20
15
10
5
0
0
0.5
1
1.5
2
Ratio NGR/O2 (Vol.)
5Nm3/h Pt
5Nm3/h Pd


CO (% Vol.) in RGR
14
12
10
8
6
4
2
0
0
0.5
1
1.5
2
Ratio NGR/O2 (Vol.)
3Nm3/h Pt
3Nm3/h Pd


CO (% Vol.) in RGF
14
12
10
8
6
4
2
0
0
0.5
1
1.5
2
Ratio NGR/O2 (Vol.)
5Nm3/h Pt
5Nm3/h Pd


UNG (% Vol.) in RGR
10
8
6
4
2
0
0
0.5
1
1.5
2
Ratio NGR/O2 (Vol.)
3Nm3/h Pt
3Nm3/h Pd

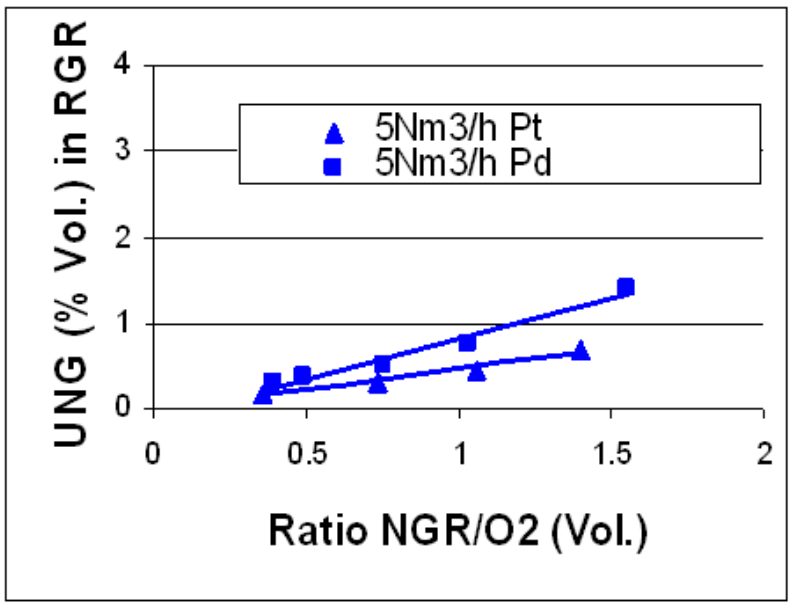

UNG (% Vol.) in RGR
4
3
2
1
0
0
0.5
1
1.5
2
Ratio NGR/O2 (Vol.)
5Nm3/h Pt
5Nm3/h Pd

Figure 7

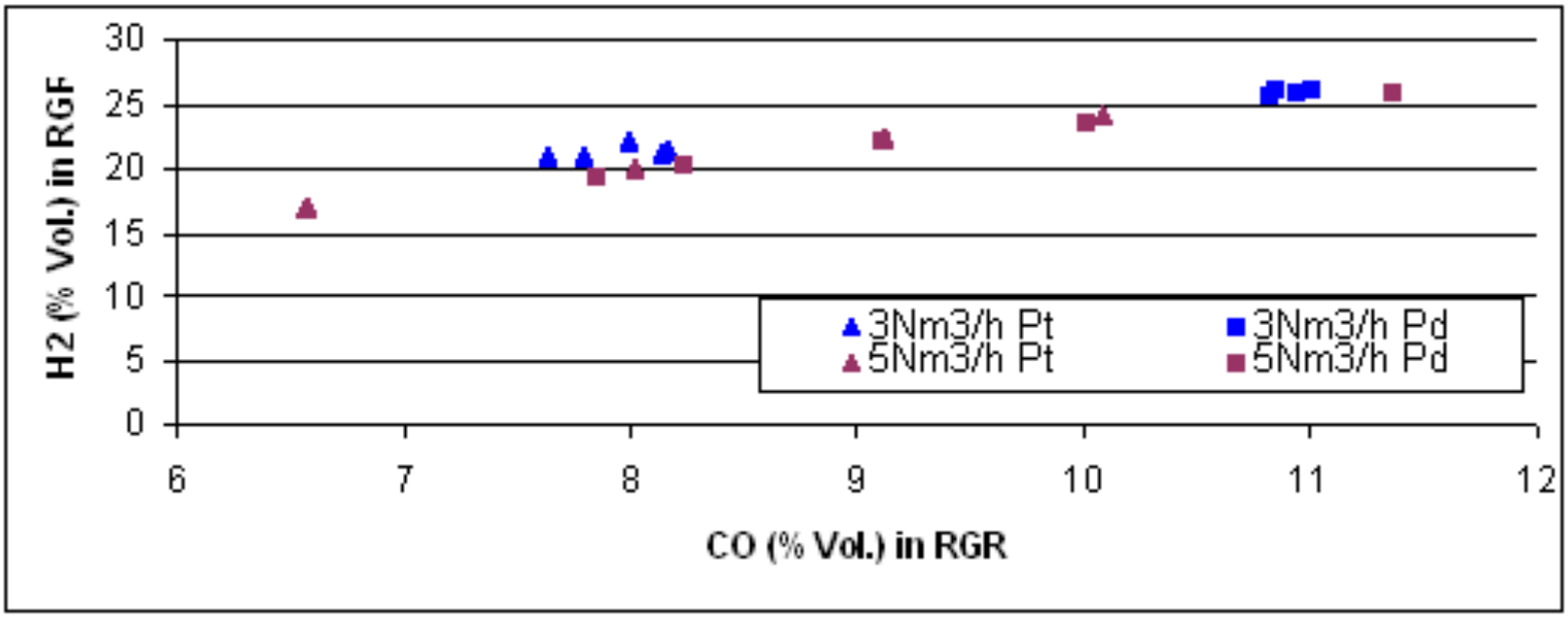
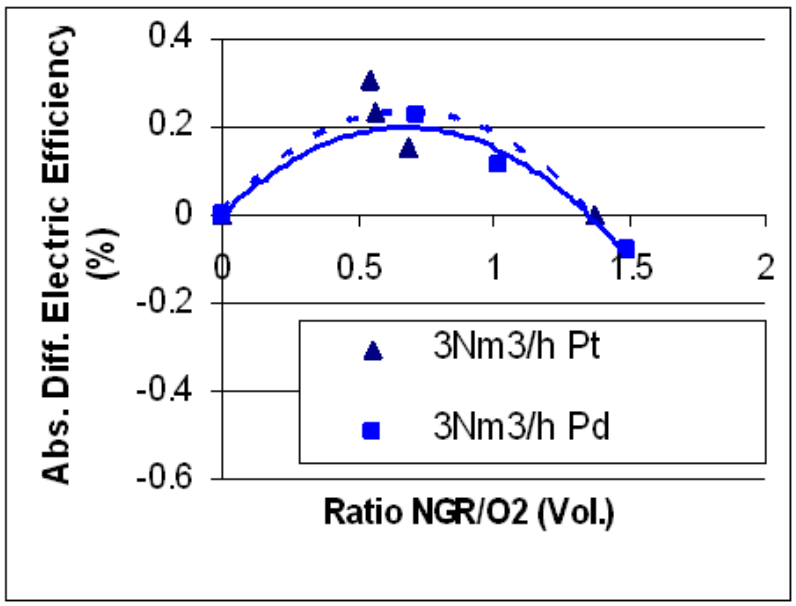

30
25
20
15
10
5
0
H2 (% Vol.) in RGF
6
7
8
9
10
11
12
CO (% Vol.) in RGR
3Nm3/h Pt
3Nm3/h Pd
5Nm3/h Pt
5Nm3/h Pd

Figure 8


Abs. Diff. Electric Efficiency (%)
0.4
0.2
0
-0.2
-0.4
-0.6
0
0.5
1
1.5
2
3Nm3/h Pt
3Nm3/h Pd
Ratio NGR/O2 (Vol.)

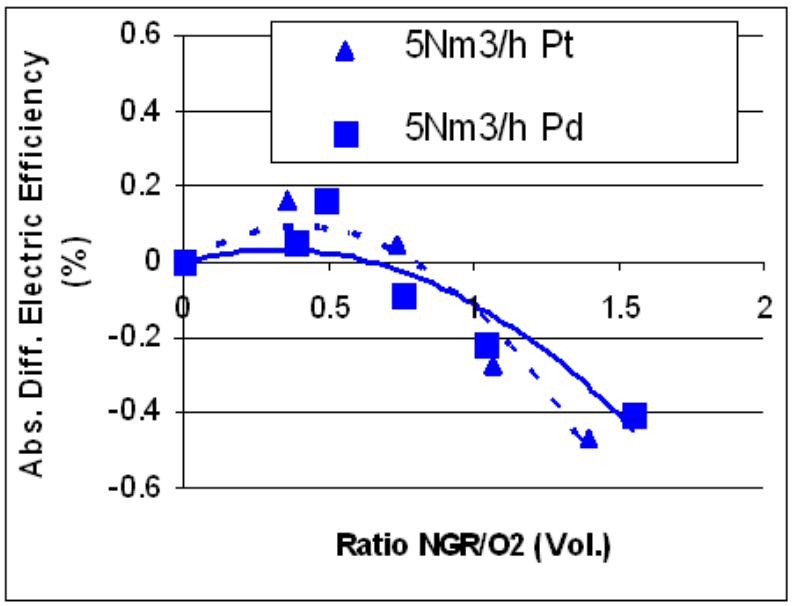

Abs. Diff. Electric Efficiency (%)
0.6
0.4
0.2
0
-0.2
-0.4
-0.6
0
0.5
1
1.5
2
5Nm3/h Pt
5Nm3/h Pd
Ratio NGR/O2 (Vol.)

Figure 9

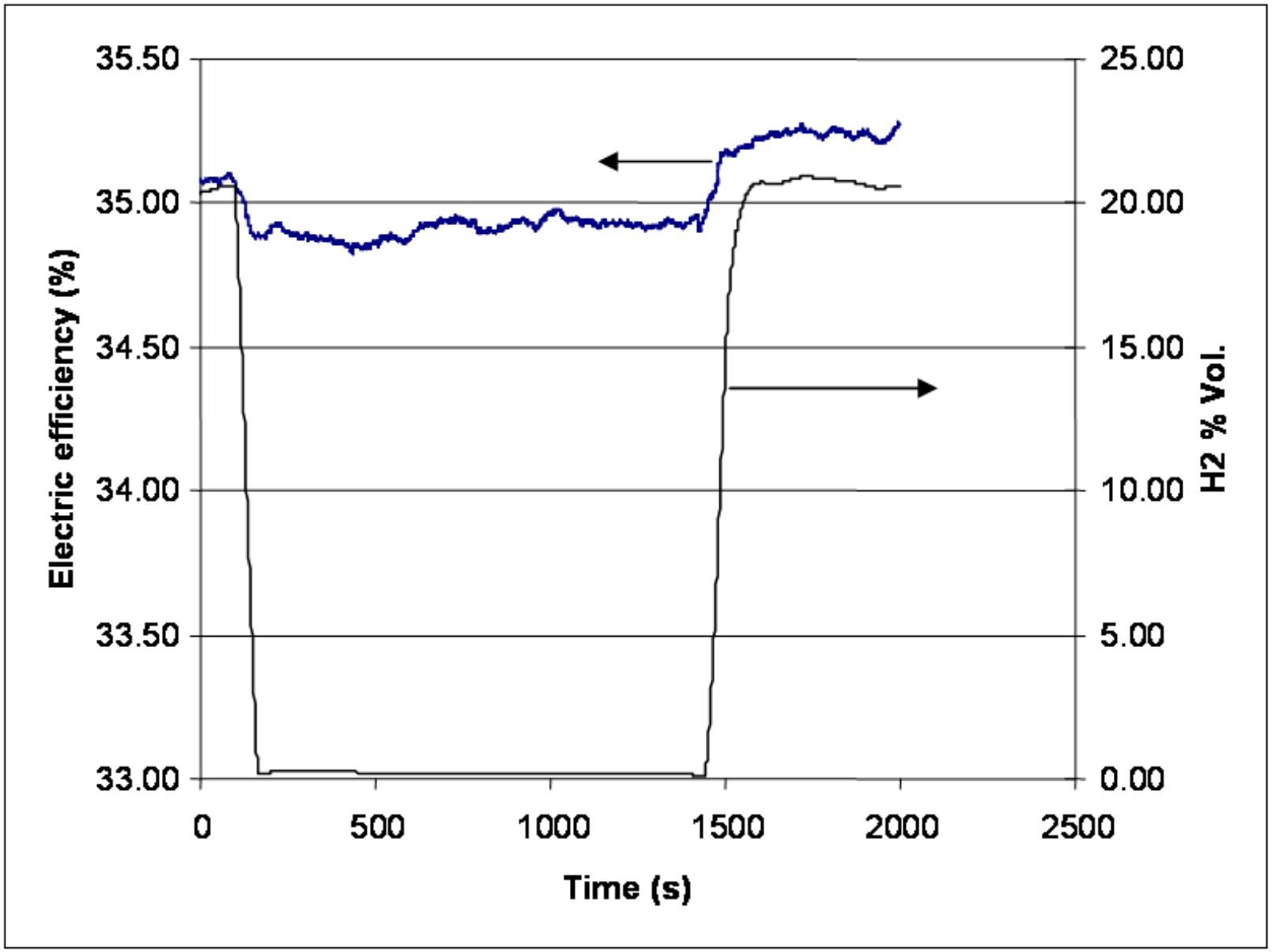
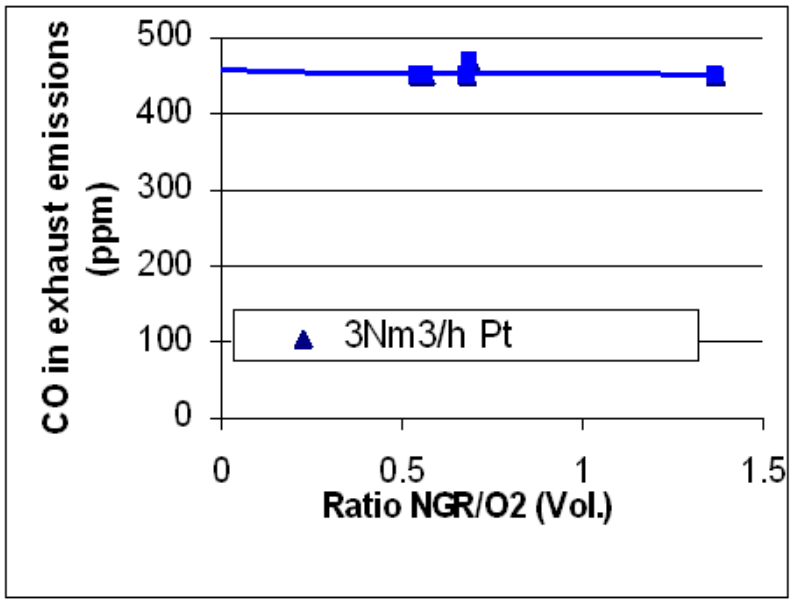
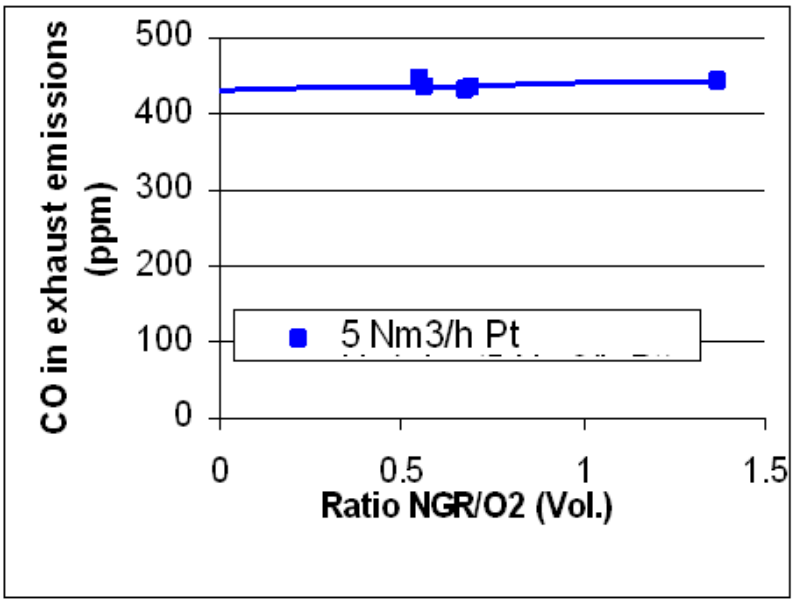
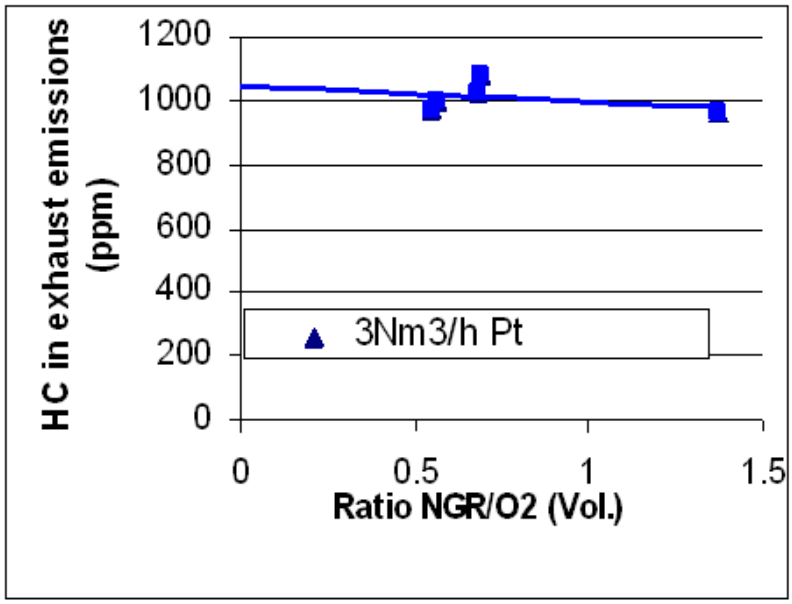

35.50
35.00
34.50
34.00
33.50
33.00
Electric efficiency (%)
25.00
20.00
15.00
10.00
5.00
0.00
H2 % Vol.
0
500
1000
1500
2000
2500
Time (s)

Figure 10


CO in exhaust emissions (ppm)
500
400
300
200
100
0
3Nm3/h Pt
0
0.5
1
1.5
Ratio NGR/O2 (Vol.)


CO in exhaust emissions (ppm)
500
400
300
200
100
0
5 Nm3/h Pt
0
0.5
1
1.5
Ratio NGR/O2 (Vol.)


HC in exhaust emissions (ppm)
1200
1000
800
600
400
200
0
3Nm3/h Pt
0
0.5
1
1.5
Ratio NGR/O2 (Vol.)

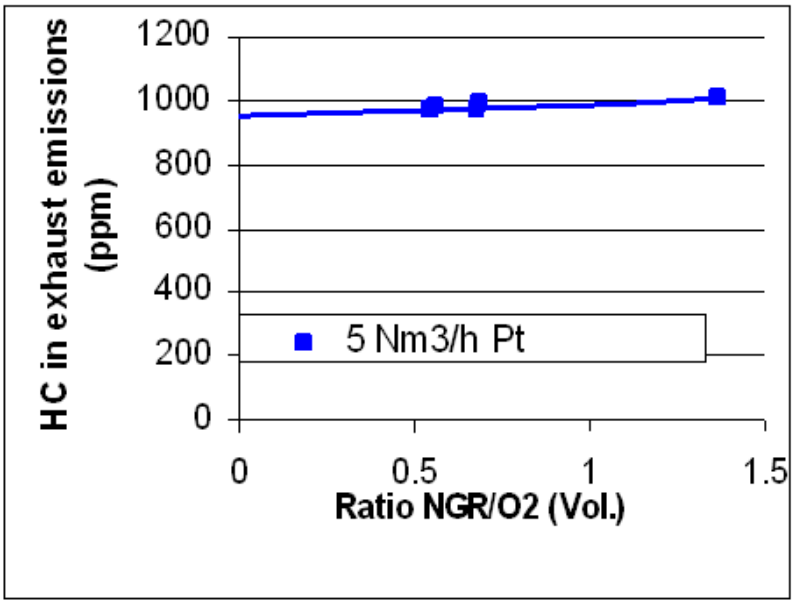

HC in exhaust emissions (ppm)
1200
1000
800
600
400
200
0
5 Nm3/h Pt
0
0.5
1
1.5
Ratio NGR/O2 (Vol.)

Figure 11