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Plasma assisted fuel reforming for on-board hydrogen rich gas production

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ABSTRACT:
Plasma assisted fuel reforming technology appears particularly attractive for automotive applications, especially regarding compactness, response time and absence of catalyst element. In 2003, Renault and CEP have initiated a research programme on this subject. A test bench allowing reformer feeding with different fuel / air / steam mixtures and coupled with a gas composition analysis system has been especially developed for this application. Preliminary results obtained under partial oxidation condition (H₂O/C: 0) have been carried out with unleaded gasoline at atmospheric pressure and around 1500 °C reactor temperature. Under these conditions, a 45 % fuel reforming efficiency was obtained (taking into account the electric power needed to generate the plasma and corresponding to H₂ and CO production). Besides, numerical models have allowed a better understanding of the reaction phenomena in the plasma reactor.

KEYWORDS : fuel reforming, non-thermal plasma, modelling, experimental results, efficiency

Introduction
Like almost all car manufacturers worldwide, Renault prepares the future, within the Renault-Nissan Alliance, by leading research programs on fuel cell technologies and their applicability to automotive use. Considering that no large scale hydrogen distribution infrastructure is available so far, and probably will not exist at medium term (before 2020), Renault focuses its research on on-board hydrogen generation from multi-fuel reforming. Whereas Nissan concentrates on long term through hydrogen on-board storage.

The main reforming technology is catalytic reforming, which has been studied by many car manufacturers and laboratories for about fifteen years and offers good efficiency. Nevertheless, this technology has some limits concerning dynamics, durability, cost and compactness. These weak points are challenges for automotive applications. In parallel with research on catalytic reforming, Renault has launched research programs on other reforming technologies.

Among alternative reforming technologies, plasma assisted fuel reforming technology appears particularly attractive for automotive applications, especially regarding response time, compactness and absence of catalyst element [1-3]. Previous works carried out at MIT based on thermal plasma have demonstrated a high efficiency in term of conversion rates. Unfortunately, these systems have shown a very poor energy conversion efficiency as a result of the high energy consumption of thermal plasmas. For this reason, non-equilibrium plasmas whose catalytic effect is effective with lower energy consumption have emerged as an interesting alternative.

The main advantages of non-equilibrium plasma reforming technology are dynamics, low energy consumption, flexibility, durability (no catalyst needed), compactness and cost. For an automotive application, plasma assisted reforming could be used complementary to catalytic reforming to ensure dynamics performance (start-up time, transients).

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After a presentation of a compact reformer and its test bench, under development in the frame of this program, this study presents some numerical and experimental results.
Experimental

A scheme of the test bench specially developed for this application is shown in Figure 1. This test bench allows the reformer feeding with different gasoline / air / steam mixtures. Gasoline flow rates up to 0.3 g/s can be achieved while O/C and H₂O/C ratios can be adjusted continuously from 0 to 1.4 and 0 to 2 respectively. Furthermore, the input temperature can be adjusted freely between ambient temperature and 800 K. Reformate gas composition is analysed by a NDIR – TCD analysis system (Rosemount, NGA 2000), which allows the continuous monitoring of H₂, CO, CO₂ and CH₄. Additional gas analyses are carried out by gas chromatography (HP 5890 serie II).

The plasma reformer is composed by a compact non-thermal arc plasma torch and a post discharge reactor. Plasma torch geometry is very similar to those encountered in classical high current DC plasma devices. An electric arc is established between a central and an annular electrode. The two concentric electrodes are separated by a high voltage insulating ceramic material. A low current - high voltage arc discharge generated between the electrodes is blown down by a high velocity gas mixture injected radially at the vicinity of the central electrode. The plasma reactor geometry has been designed to further a high plasma homogeneity and an efficient reagents mixing.

Figure 1: Overall scheme of the plasma reformer and its test bench and view of the test bench.
The generation of a low current - high voltage arc discharge is obtained using a power supply based on a resonant converter technology specially developed for this application. A 15,000 V maximum voltage can be achieved while arc current is limited to 660 mA. Contrary to high voltage transformers currently used for similar applications, this power supply provides the continuous control of the arc current with a high accuracy in the range 200-600 mA. Depending on the conditions (gas flow rate, current), the regime of the discharge can vary from streamer over gliding arc to continuous discharge [4]. Electrical signals are analysed by a 2 channels digital oscilloscope (HP 54615 B). Voltage measurement is performed using a 1:1000 probe (Elditest, GE3830) while discharge current measurement is carried out with a hall effect current probe (PR 30: LEM).

The post discharge reactor is located downstream the plasma torch. The axial temperature along the chamber is measured by four thermocouples. The reformer pressure is controlled by a valve located at the reformer output. All operating parameters are collected by a data acquisition system (HP 34970 A) and recorded in a PC.

**Modelling**

**PSR Model**

In a first model, it is assumed that the plasma uniquely acts as a power source. The n-Octane reforming is studied by applying a PSR (Perfectly Stirred Reactor) approximation with a kinetic reaction mechanism developed in the frame of combustion [5].

By using this approach, the influence of the main processing parameters composition (O/C and H$_2$O/C ratios), inlet temperature, pressure, plasma power and reactor volume, can easily be described. This study only consider the composition and the reactor volume influence.

**Plasma reformer Model**

In a more sophisticated model, a first approach to take into account the catalytic effect of the plasma is developed. The model, adapted from [6], considers that only a fraction of the gas flow passes through the arc zone. Here the gas composition changes and it is calculated using a PSR sub model. Once this fraction leaves the arc region, it is assumed that it is perfectly mixed with the remaining gas. At this time, the composition of the mixture is calculated by using a piston (1D) flow reactor sub model. As the gas fraction passing through the arc zone can hardly be estimated, a sensitivity study on this parameter has been carried out.
Results

PSR Model results

Figure 2 and 3 present results obtained with the PSR model. Chemical efficiency versus O/C ratio and H₂O/C ratio are plotted for different reactor volumes and compared with the thermodynamic value.

Figure 2 shows that the higher the O/C ratio, the higher the temperature and the faster the kinetic. Thus, kinetic data goes closer to thermodynamic values. The temperature increases due to exothermal reaction of partial oxidation.

Figure 3 shows that maximum values of conversion efficiency are obtained for the same H₂O/C ratio (equal to 0.4). This H₂O/C value corresponds to the part of hydrocarbon that isn’t consumed by the oxygen (O/C ratio) and that could possibly react with H₂O. This means that partial oxidation kinetic is faster than steam reforming one. Moreover, the higher the H₂O/C, the lower the temperature, and the lower the kinetic. This temperature decreases as a result of both the endothermal steam reforming reaction and the steam dilution.
In addition, the efficiency increases with the reactor volume since the residence time increases.

**Plasma reformer Model results compared to experiment results**

The experimental analysis of the compact plasma reformer is currently under way. This analysis will allow to study the influence of the main processing parameters: flow rates, pressure, inlet composition, power of the plasma,... Preliminary results obtained under partial oxidation condition (H2O/C : 0) have been carried out with unleaded gasoline at atmospheric pressure and around 1500 °C reactor temperature. Under these conditions, a 45 % fuel reforming efficiency was obtained. This value takes into account the electric power needed to generate the plasma and corresponds to H2 and CO production (assuming that CO could be totally converted into H2 in a post water gas shift reactor). Molar fractions obtained at the reactor output were (dry composition): CO : 18 %, H2 : 17 %, CO2 : 4 % and CH4 : 2 %. These results appear very promising since they have been obtained before real process optimisation.

![Reformate dry gas composition](image)

**Figure 2 : Conversion efficiency versus H2O/C ratio for different reactor volume obtained with model 1 (O/C : 0.6, T_{inlet} : 400 K, Q_{fuel} : 0.1 g/s, P : 1000 W)**
Conclusion

A new plasma assisted compact reformer for on-board hydrogen production has been developed and is currently under characterisation. This technology results from a research collaboration programme between Renault and CEP. Experimental results obtained before process optimisation seem very encouraging. In parallel, an important effort for a better understanding of the reaction phenomena has been carried out through different numerical models.

References