

PSFC-JA-05-22

**Plasmatron Fuel Reformer Development and
Internal Combustion Engine Vehicle Applications**

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August 31, 2005

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Paper presented at the
Diesel Engine Emission Reduction (DEER) Workshop, 2004
Coronado CA, August 29-September 2, 2004

Supported by Department of Energy, Office of FreedomCar and Vehicle Technologies

PLASMATRON FUEL REFORMER DEVELOPMENT AND INTERNAL COMBUSTION ENGINE VEHICLE APPLICATIONS

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ABSTRACT

Onboard generation of hydrogen-rich gas can be used in a number of applications to improve environmental quality and reduce petroleum consumption of internal combustion engine vehicles. Plasmatron fuel reformer technology is being developed at MIT as means of practical on-board production of hydrogen-rich gas from a variety of fuels. The device is based on the use of a special low current, low power volumetric discharge in very rich fuel/air mixtures. Although the process is slightly exothermic, system advantages overwhelm the slight inefficiency of the process. In this paper, recent progress on plasmatron fuel reformer and applications are described.

In 2003, a plasmatron fuel reformer was used by ArvinMeritor in a successful test on a bus of a two leg NO_x trap exhaust aftertreatment system regenerated with hydrogen-rich gas [1]. The input diesel fuel flow rate to the plasmatron fuel reformer was 0.8 g/s. In 2004 the input fuel flow rate of a diesel plasmatron fuel reformer at MIT was operated with an increased diesel fuel flow rate of 2 g/s, corresponding to 80 kW of fuel reformat. This reformer produced about 1.5 l/s of H₂ without the use of a reforming catalyst for additional hydrogen generation. This type of higher flow rate operation could facilitate quick regeneration of single-leg NO_x trap systems aftertreatment.

Reformation tests of renewable fuels have been carried out. Vegetable oils, including soy and canola oils, have been efficiently reformed, with no soot production at input rates of 0.5 g/s. Ethanol tests have been carried out with. Both non-catalytic and catalytic (with a catalyst downstream from a homogeneous reforming zone) have been studied. Onboard conversion of biofuels into hydrogen rich gas opens up a range of opportunities for reducing petroleum consumption.

The plasmatron response has been determined using real time gas analysis techniques. For cold start, the response of a plasmatron fuel reformer using homogeneous reforming was faster than that from a plasmatron fuel reformer that included a catalyst. In addition, hydrogen production during warm start has also been studied. This mode of operation could be particularly relevant to some aftertreatment applications, where the hydrogen rich gas needs to be produced in short, frequent pulses.

Concepts have been developed for optimal regeneration of Diesel Particulate Filters, using hydrogen rich gas. The goal is the simplification of the aftertreatment system, increasing reliability, fuel penalty minimization, and minimization of fuel reformer system requirements

INTRODUCTION

In order to meet stringent US emissions regulations for 2007-2010 diesel vehicle model years, after-treatment technology is being developed. Both particulate and NO_x emissions after-treatment technology will be necessary, as it seems that engine in-cylinder techniques alone will be unable to meet the regulations.

MIT, in collaboration with ArvinMeritor, has been developing plasmatron fuel converter for onboard applications, including engine fuel conditioning as well as diesel engine aftertreatment [1,2]. The MIT technology has been licensed by ArvinMeritor. Description of ArvinMeritor's effort in diesel exhaust aftertreatment can be found in a paper by Khadya [3].

In addition to diesel fuel reformation, the present paper describes efforts in reforming renewable fuels. The use of reformat from either diesel or renewable fuels for diesel and diesel-like engines is explored. The purpose of the present work is to develop hydrogen based technology for addressing after-treatment issues [4-6]. In this paper recent developments in both the technology for on-board generation of hydrogen as

well as applications to Homogeneous Charge Compression Ignition (HCCI) engines will be discussed. The possible use of reformat for regeneration of Diesel Particulate Filter (DPF) will also be described. This application will be experimentally investigated at MIT in the near future,

PLASMATRON TECHNOLOGY

Plasmatron fuel converter technology consists in the use of a plasma discharge for the initiation of reformation process. Proper placement of the CW discharge is used in a flowing air/fuel mixture that is very rich. Proper preparation of the fuel and fuel/air mixture is required for optimal operation.

The process is schematically shown in Figure 1. The flowing air/fuel mixture encounters a non-equilibrium plasma discharge, operating at about a thousand Volts and at currents on the order of hundreds of mA. The plasma region is followed by a homogeneous reactor where the volume reactions started in the plasma reactor result in the generation of a reformat gas consisting of hydrogen, CO, light hydrocarbons and small amounts of products of combustion, at relatively high temperature (~1000 °C). As indicated in Figure 1, an optional catalyst is used for the further processing of the hydrogen rich gas, further processing the light hydrocarbons into additional hydrogen and CO.

The plasmatron fuel reformer technology is attractive because of capability for fast response, elimination or relaxation of catalyst requirements, and fuel flexibility. Fast response is needed for onboard applications in SI engines, where on-demand, wide dynamic range hydrogen rich gas requirements are stringent, as well as in aftertreatment applications, where short burst of hydrogen rich gas are needed. Good hydrogen yields (defined as the ratio between the hydrogen gas in the reformat to the hydrogen in the hydrocarbon fuel) have been achieved without the use of catalysts (about 40%, with ~70% to H₂ and light hydrocarbons). The use of a catalyst downstream from the plasma region increases the hydrogen yield to ~70%. Experiments have been conducted with gasoline, diesel, ethanol and hard-to-reform bio-oils.

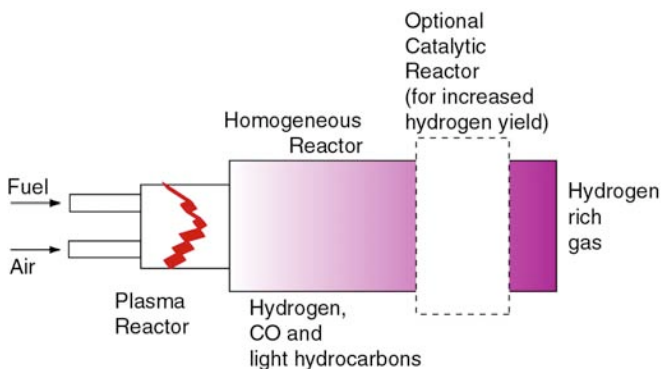


Figure 1. Schematic of plasmatron fuel reformer.

The plasmatron reformer used in the biofuel conversion experiments is a modification of a plasmatron fuel reformer that had been optimized for diesel fuel operation¹. The unit was operated mainly at 700 W of electrical power. Limited

tests were conducted at 200 W. The power supplies were operated as constant current sources operating between 200 kHz and 260 kHz.

Reforming experiments at different oxygen-to-carbon (O/C) ratios were carried out. In the case of ethanol, some of the oxygen required for the reformation is provided by the fuel itself. In this case, the O/C ratio includes this oxygen. The free oxygen was provided by supplying compressed air from building compressed air supply. The bio-oils used in the experiments were commercially available oils

Figure 2 depicts the experimental setup used to reform diesel fuel, vegetable oils and ethanol, while Figure 3 shows a photograph of the plasmatron fuel converter used in the experiments. The plasmatron was mounted on top of a steel reaction chamber that allows the homogeneous partial oxidation reactions to take place with enough residence time. The dimensions of this chamber are 5 cm diameter by 12.5 cm long. The products of reaction from this chamber go into a 5 cm diameter by 17.5-cm long chamber packed with nickel-based catalyst on an alumina substrate. The chamber is thermally insulated by a half-inch thermal insulation made of porous alumina ceramic.

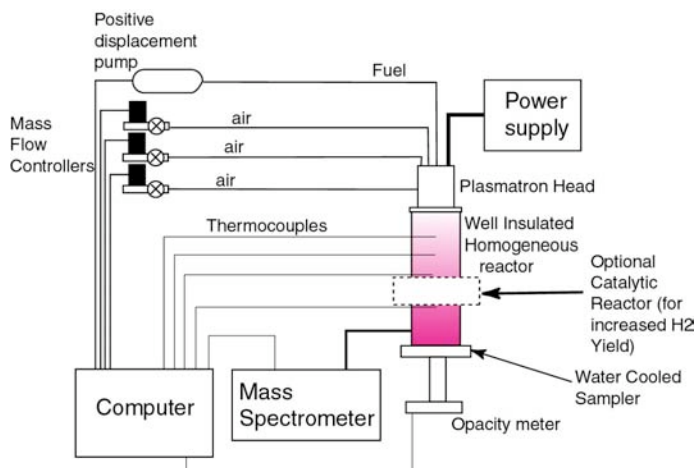


Figure 2. Experimental setup.

Two sample ports are used for sampling the reformat gases. The first one is located on the reaction chamber immediately upstream from the catalyst and the second one is located at the exhaust, downstream from the catalyst chamber. Both sampling lines are water cooled. The temperature inside the reactor is measured by a thermocouple located inside the catalyst.

Calibrated mass flow controllers are used to control the multiple air inputs into the plasmatron. The system is controlled by a LabView software developed in our laboratory. The software continuously acquires and stores several parameters such as temperature, air and fuel flow rates during operation.

The fuel is injected from the top of the plasmatron through a commercially available nozzle. Directly downstream from the fuel nozzle there is an orifice. The high velocity of the air downstream from this orifice further atomizes the fuel into a fine mist.

To prevent the fuel from striking the solid surfaces inside the plasmatron and produce soot by pyrolysis, wall air is

introduced through an annular gap between the electrode and the atomization air plug. This wall air creates a sheath along the inner walls of the plasmatron and prevents the fuel from hitting the wall. A third air stream, the plasma air, is introduced with a large tangential component in the gap between the two cylindrical electrodes where the electric discharge takes place. The tangential motion of the plasma air prevents the plasma from attaching to one spot on the electrodes, by moving the discharge and its roots. The plasma air pushes the discharge into the main volume of the plasmatron, producing a discharge that in an average sense fills this region.

Samples from the reformat gases are drawn into syringes and analyzed using an MTI-2000 gas chromatograph (GC). The unit has a molecular sieve column for analyzing hydrogen, oxygen, nitrogen, methane, and carbon monoxide, and a second column that can analyze carbon dioxide, C₂H₂, C₂H₄, and C₂H₆, and other heavier hydrocarbons. A WAGNER opacity meter was used to monitor the production of soot. Water is not monitored.

A mass spectrometer was used in order to characterize the dynamic response of the plasmatron fuel converter. The time response (10-90%) of the flow meters was about 100 ms. However, because of long lines between the flow meters and the experimental setup, the time response of the gas system was considerable longer. Efforts will be undertaken in the future to decrease the volumes between the gas flow meters and the plasmatron.

The measurement system consisted of a 200 μm ID capillary 1.6 m long directly introduced into the flowing reformat, in order to minimize time response of the measurement. A novel scheme was developed that allowed qualitative measurements of flow rates, using Ar as the benchmark. Since the flow rates of air are known, and air contains substantial amounts of Ar (about 1%), the flow rate of Ar in the reformat is known. By taking ratios between the signals of the gases of interest and Ar, the flow rates of the gases of interest can be determined. This avoids problems of absolute calibration of the mass spectrometer, which is subject to variations due to temperature effects, partial blockages of the capillary, and other environmental effects. The setup can be used for determining concentration with appropriate choice of Ar concentration in the reformat. Alternatively, the N⁺ ion can be used instead of Ar, but our preliminary results indicate that lower noise is obtained with Ar as the benchmark. The device was optimized for making measurements of 10 ions every 200 ms. A paper is being prepared that describes the instrument development described above.

In order to increase the hydrogen concentration in the reformat, a catalyst can be placed downstream from the homogeneous reforming region. The homogeneous stage of the plasmatron fuel reformer converts the liquid hydrocarbons into hydrogen, CO and light hydrocarbons, plus a small amount of carbon dioxide and water. The catalyst is useful in converting light hydrocarbons into additional hydrogen. The corn and soybean catalytic experiments were done with a volume of 85 cm³ of catalyst, whereas only 25 cm³ of catalyst were used during the canola oil experiments.

At present, goals for the diesel plasmatron reformer development includes increased flow rates, operational and

configuration optimization and increased simplicity, among other goals.

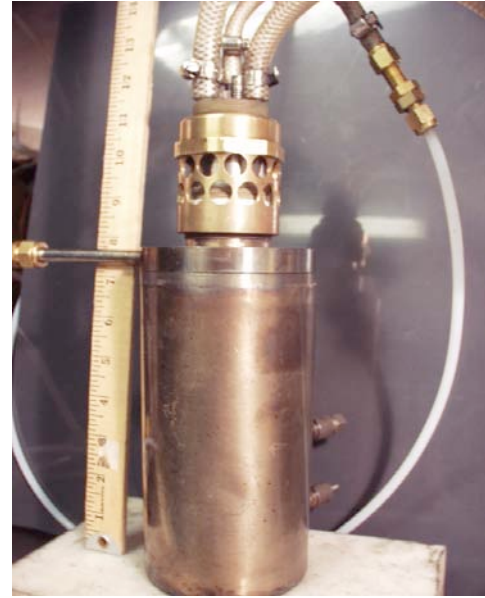


Figure 3. Compact low power plasmatron fuel converter

DIESEL REFORMATION

In 2003, a plasmatron fuel reformer was used by ArvinMeritor in a successful test on a bus of a two-leg NO_x trap exhaust aftertreatment system regenerated with hydrogen-rich gas [1]. The input diesel fuel flow rate to the plasmatron fuel reformer was 0.8 g/s.

Table 1.

Experimental Results of Homogeneous Diesel Reforming		
Electric power	W	200
O/C		1.2
Diesel flow rate	g/s	0.8
Corresponding chemical power	kW	36
Reformat composition (vol %)		
H ₂		7.6
O ₂		1.3
N ₂		64.0
CH ₄		2.4
CO		13.0
CO ₂		4.4
C ₂ H ₄		2.2
C ₂ H ₂		0.0
H ₂ O		7.1
Energy efficiency to hydrogen, CO and light HC		65%
H ₂ flow rate	l/min (STP)	20
Soot (opacity meter)		0.0%

In 2004 the input fuel flow rate of a diesel plasmatron fuel reformer at MIT was operated with an increased diesel fuel flow rate of 2 g/s, corresponding to 80 kW of fuel reformat. This reformer produced about 1.5 l/s of H₂ without the use of a reforming catalyst for additional hydrogen generation. This

type of higher flow rate operation can facilitate quick regeneration of single-leg NO_x trap systems aftertreatment.

The plasmatron fuel converter has been further developed for use with diesel fuel. Results from the plasmatron diesel fuel converter are shown in Table 1. The goal of reformation of diesel by the plasmatron fuel converter is the conversion of the heavy diesel compounds into hydrogen, carbon monoxide, and light hydrocarbons for use in after-treatment applications. The diesel reformat produced by the plasmatron fuel converter has little residual oxygen and little or no soot. The diesel plasmatron can be cycled on and off quickly (~ 3-5 sec). For diesel after-treatment applications the dynamic performance of the plasmatron fuel reformer is important because of the low duty cycle of operation. For NO_x catalyst regeneration applications, the plasmatron may be turned on for a few seconds every half minute or so. For diesel particulate filter applications, the plasmatron could well be operated for a few minutes every few hours resulting in much smaller duty cycles.



Figure 4. Gen H plasmatron developed at ARM.

Hydrogen is a powerful reductant, and its use for the regeneration of NO_x traps has been researched in catalyst reactor labs. The goal of the work presented next is to determine the advantages of H₂ assisted NO_x trap regeneration, using the plasmatron fuel reformer as the source of on-board hydrogen. Tests at ArvinMeritor have demonstrated the

advantages of using hydrogen rich gas for regeneration and desulfation of NO_x traps. Substantially higher regeneration of NO_x traps with hydrogen can be obtained with hydrogen rich gas. In contrast to use of diesel fuel, adequate regeneration can be obtained at low exhaust temperatures (in the 150-200°C range). In addition, lower fuel penalty is observed when comparing hydrogen-rich gas regeneration to diesel fuel regeneration. The temperatures required for desulfation are also substantially decreased. Additional information on the tests and implementation of the plasmatron technology by ArvinMeritor is presented in a paper by Khadya [3]. Figure 4 shows a Fuel Reformer developed at ArvinMeritor.

BIOFUEL REFORMATION

Reformation tests of renewable fuels have been carried out. Vegetable oils, including soy and canola oils, have been efficiently reformed, with no soot production at input rates of 0.5 g/s. Ethanol tests have been carried out with. Results from both non-catalytic and catalytic (with a catalyst downstream from a homogeneous reforming zone) will be presented. Onboard conversion of biofuels into hydrogen rich gas opens up a range of opportunities for reducing petroleum consumption.

BIO OILS

Figure 5 shows results for hydrogen yields from the reformation of canola, corn, and soybean oils as a function of O/C ratio for catalytic and homogeneous (non-catalytic) reforming. The hydrogen yield is defined as the ratio of the mass flow rate of hydrogen in the reformat gases to the mass flow rate of hydrogen in the fuel. The bio-oil flow rate was 0.4 g/s, which corresponds to about 17 kW of chemical power.

The hydrogen yield for the reformation of corn and soybean oils is very similar in the presence of a catalyst. The difference between the two falls within the error bar due to sampling and GC analysis uncertainties.

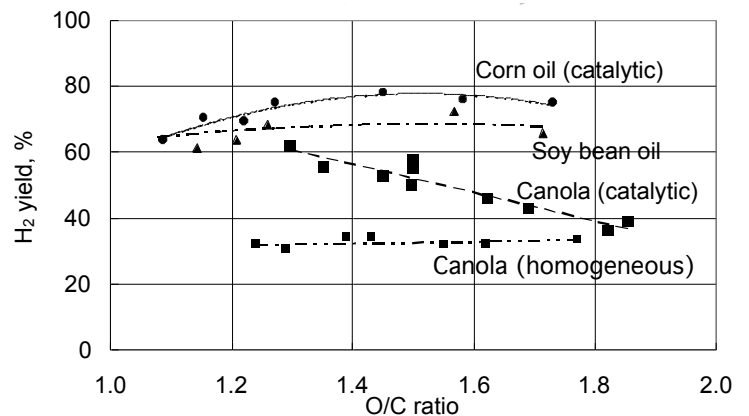


Figure 5. Hydrogen yield for catalytic reformation of corn, canola and soybean oil.

On the other hand, the hydrogen yield for canola oil in the presence of a catalyst appears to be lower than for corn and

soybean oils, especially at the higher values of O/C. This may be due to the use of less catalyst in the case of canola, or to issues with the air-fuel injection (for example, misalignments of the injector where the oil spray hit the walls). In the case of corn and soybean oils, the hydrogen yield peaks at an O/C ratio close to 1.5; this is due to the fact that some of the fuel needs to be combusted in order to provide the heat necessary to drive the partial oxidation reaction.

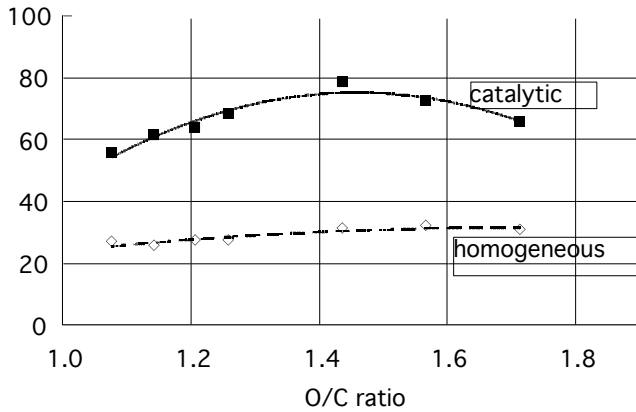


Figure 6. Hydrogen yield as a function of O/C ratio for partial oxidation reformation of soybean oil.

Comparison between homogeneous (non-catalytic) and catalytic reforming was carried out for the case of corn, canola and soybean oil. Figure 6 shows the effect of catalyst on the hydrogen yield, for a fixed soybean oil flow rate of 0.37 g/s. The hydrogen yield is the ratio of hydrogen in the form of H₂ to the hydrogen in the biofuel. For homogeneous reformation of soybean oil, the hydrogen yields are about 30%, while the yield increases to about 80% in the presence of a catalyst. The peak yield occurs at O/C ~ 1.5. The difference between the yield of 80% and the ideal yield of 100% is due primarily to losses of hydrogen resulting from full oxidation into water. For homogeneous reformation, the ratio of the heating value of the hydrogen, CO and light hydrocarbons byproducts to the heating value of the fuel was typically between 60 and 70%.

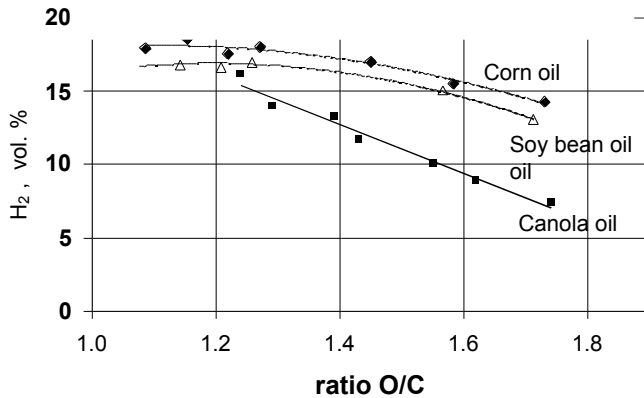


Figure 7. Hydrogen concentration (vol.) for catalytic reformation of corn, soybean and canola oils.

The composition of the gas reformation upstream and downstream the catalyst for soybean oil for an O/C ratio of 1.08 is shown in Table 2. The plasmatron converts homogeneously the high hydrocarbons into hydrogen, carbon monoxide and light hydrocarbons, with minimal soot production and eliminating all free oxygen. The catalyst then takes the oxygen-free plasmatron gas and basically doubles the hydrogen yield performing CO₂ and possibly steam reforming (notice that the CO₂ concentration is decreased downstream the catalyst; water, on the other hand, is not monitored).

Table 2.
Reformate composition of unrefined soybean oil
Flow rate:0.37 g/s (16 kW) with O/C = 1.08

	Homogeneous vol %	Catalytic vol %
H ₂	8.1	15.5
CO	11.1	16.6
CO ₂	6.7	5.0
N ₂	57	53
CH ₄	1.2	1.0
C ₂ H ₄	5.1	2.8
C ₂ H ₂	0.1	0.0
O ₂	0.6	0.6
H ₂ O*	9.8	5.3

*Estimated from hydrogen and oxygen balances

It should be stressed that the performance of the system was not optimized, and higher hydrogen yields could be possible by converting the substantial amounts of C₂'s present in the gas downstream from the catalyst.

For all the experiments performed with veggie oils, the opacity was 0.1, which is the sensitivity limit of the instrument and which indicates that at most very small amounts of soot were produced.

PLASMATRON REFORMATION OF ETHANOL

Test with ethanol were conducting using the same experimental protocol and setup use for bio-oils. Both homogeneous (non-catalytic) and catalytic reforming have been explored.

Transients results, with the plasmatron fuel converter at room temperature, were carried out. The startup and steady state composition of the reformate were investigated using the mass spectrometer described above.

Preliminary investigation of startup optimization through variation of O/C ratio were explored.

Figure 8 shows the concentration (vol. %) of hydrogen, CO, ethane and methane as a function of time for the case of homogeneous reforming (without the use of a catalyst). Hydrogen concentration is about 6% right at start, increasing slowly to about 12% (in 150 seconds). These results were obtained using a discharge power of 200 W with a ethanol flow rate of 0.9 g/s, corresponding to 26 kW of heating power. The

O/C was ramped from an initial value of 2.3 at start-up to 1.73 at 12 s. Although there was some effort in trying different ramp rates, the startup scenario of Figure 8 is not fully optimized and it may be possible to further increase both the fast transient during the initial 12 s and the slow ramp after 12 s.

Figure 9 shows the yields to hydrogen (ratio of hydrogen gas in the reformat to hydrogen in the fuel), as well as the hydrogen and light hydrocarbon yield (ratio of hydrogen in H₂ and light hydrocarbons in the exhaust to hydrogen in the fuel) for the same conditions as Figure 8. for the Figure

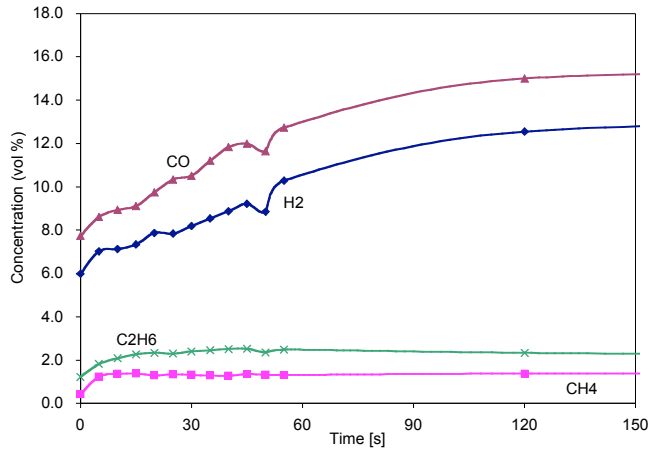


Figure 8. Composition of the reformat as a function of time for homogeneous reforming, with variable O/C ratio during the first 12 seconds.

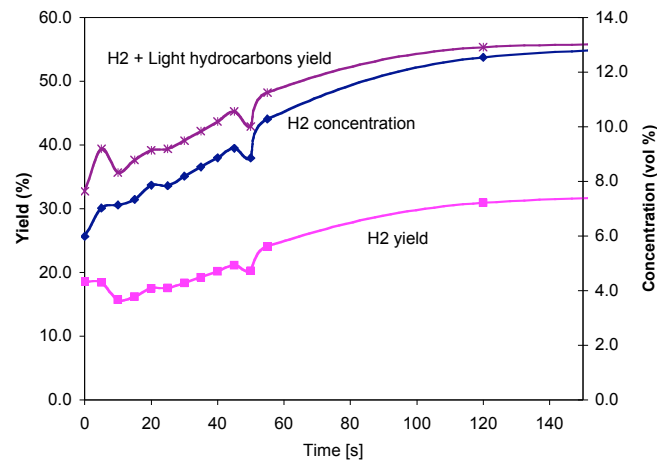


Figure 9. Same a Figure 8 for hydrogen and hydrogen/light hydrocarbon yields.

In addition to homogeneous reforming, combined homogeneous and catalytic reforming was explored as means of increasing the hydrogen yield. These results are presented in Figure 10. The electrical power in the discharge is the same as for homogeneous reforming, as well as the ethanol flow rate. The ramp of the O/C ratio, however, is slightly different, as the final O/C ratio in the case of catalytic reforming is lower than that for homogeneous reforming. In the case of catalytic reforming shown in Figure 9, the O/C ratio is ramped from O/C

~ 2.3 at startup to O/C ~ 1.56 at 12 s. The hydrogen concentration and yields are higher than in the case of homogeneous reforming shown in Figures 8 and 9, even at startup, with an initial H₂ concentration of 10%. In addition, the steady state value of the hydrogen concentration and yields are substantially higher than in the case of homogeneous reforming. In addition to higher yields, the rate to achieve high conversion significantly higher than in the homogeneous case, reaching near steady state at the end of the 12 s ramp-up. The energy efficiency (ratio of the heating value of the reformat to the heating value of the ethanol), is greater than 70% at steady state.

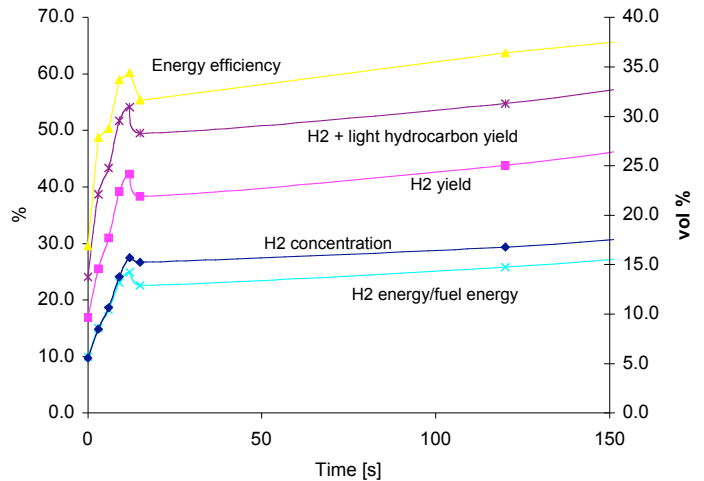


Figure 10. Plasma-catalytic reformation of ethanol.

HYDROGEN RICH GAS ENHANCED DPF REGENERATION

Options of using reformat from a plasmatron fuel reformer for the controlled regeneration of DPFs are being explored.

Hydrogen rich gas could provide important advantages for controlled regeneration of DPF. The reformat can provide clean burning fuel to initiate and control burnup of soot in trap during regeneration. Alternatively, the use of hydrogen rich gas may results in DPF regeneration at lower soot levels. This regeneration scheme would result in more frequent regenerations, but with the advantage of decreased chance of large uncontrolled regeneration that could negatively impact the performance of the trap, thus resulting in increased reliability and longevity.

Active techniques for the regeneration of diesel particulate filters use a burner to achieve the temperatures at the catalyst required for filter regeneration. Plasmatron technology can be used as a fuel igniter as well as means to produce hydrogen rich gas. Plasmatron technology can serve as a powerful igniter and thus is attractive because it is possible to combust the diesel fuel using oxygen present in the diesel exhaust, even at conditions when the engine is operating at high load. The fuel combustion can take place even under conditions where the free oxygen to fuel ratio is larger than unity, *i.e.*, lean burn conditions in the plasmatron.

The advantage of combusting the fuel in the exhaust is that it is possible to achieve combustion while minimizing the amount of extra air introduced into the system. The hydrogen

rich gas with exhaust gas as oxidizer results in a net decrease of the amount of free oxygen in the exhaust, decreasing the combustion rate of the soot on the DPF and decreasing the possibility of uncontrolled regeneration. This technology is attractive because the plasmatron hardware may eventually already exist on-board the vehicle for regeneration of NO_x absorber traps.

The plasmatron fuel reformer can be used as a powerful combustor, operating with exhaust as the oxidizer. There is thermal output from the combustor, but no hydrogen or light hydrocarbons. Alternatively, the reformer operates under conditions close to partial oxidation (free oxygen to carbon ratio of 1), producing hydrogen, CO, light hydrocarbons and limited thermal output (~ 1/5 of the thermal output is the fuel is combusted). These two are extremes, with the plasmatron operating anywhere in between, generating both easily combustible gases as well as thermal output.

Two options of location of the DPF and NO_x trap exist. The DPF can be upstream from the NO_x trap, or the other way around. Integration of the system is required to determine the optimal configuration. Care must be taken that the temperature required for regeneration of the DPF does not damage the NO_x trap, as well as the issues having to do with desulfation of the NO_x trap.

RESULTS SUMMARY

- Plasmatron fuel converters have been shown to be good sources of hydrogen rich gas for regeneration of NO_x traps
- Bio-oils (even unrefined oils) and ethanol can be efficiently and cleanly reformed into hydrogen rich gas.
- Increased hydrogen conversion and hydrogen using plasma catalysis has been demonstrated for diesel, bio-oils and ethanol.
- Plasmatron fuel converters offer advantages for regeneration of DPF.

FUTURE EFFORTS

- Increase hydrogen rich flow rate for online regeneration of NO_x traps.
- Tests of DPF regeneration using hydrogen rich gas
- Improved plasmatron fuel converter design with improved ease of manufacturing

Nomenclature

Reformate – the gas mixture resulting from the partial oxidation of fuel

O/C – ratio of oxygen atoms to carbon atoms in the reformer air-fuel mixture prior to processing

NO_x – symbol for the variable mixture of NO and NO₂ present in engine exhaust

ACKNOWLEDGMENTS

This work was supported by US Department of Energy, Office of Freedom CAR and Vehicle Technology. We thank Dr. S. Diamond for his support and encouragement of this work.

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♣ Work supported by the Office of Freedom car and Vehicle Technologies, US Department of Energy.