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**Aftertreatment of Diesel Vehicle Emissions
Using
Compact Plasmatron Fuel Converter-Catalyst
Systems**

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Abstract

Improvements in diesel engine aftertreatment technology are needed to meet future environmental quality goals. Systems using recently developed compact plasmatron fuel converters in conjunction with aftertreatment catalysts could provide new opportunities for obtaining significant pollution reductions. Use of compact plasmatron reformers to produce hydrogen-rich gas for the regeneration of NO_x absorber/adsorbers and particulate traps could provide significant advantages. Catalyst requirements are determined and overall system characteristics are evaluated. The options made possible by the use of the on-board hydrogen generation for the regeneration of catalysts are briefly described.

I. Introduction

Diesel engines are presently the most efficient vehicular propulsion systems. Heavy trucks and buses are powered almost exclusively by diesel engines worldwide. In Europe, diesel powered cars have been increasingly popular. The high reliability of these engines is an additional attractive feature. The diesel engine may become even more widely used in the future. However, significant further progress in diesel emission control is needed. Diesel particulates and nitrogen oxides, the two most troublesome components of diesel exhaust emissions, have adverse impact on urban air. The new 2004 emission standards in the U.S. (to be implemented in 2002 after the consent decree between the manufacturers and the EPA) as well as the tightening regulations in Europe and East Asia reflect the growing concern with diesel emissions. Recently developed compact plasmatron fuel converters have features that are suitable for onboard production of hydrogen for aftertreatment applications. Systems that use these devices in conjunction with aftertreatment catalysts have the potential to significantly improve prospects for reduction of diesel emissions.

Compact plasmatron fuel converters have been shown to efficiently reform natural gas [Bromberg1], gasoline [Bromberg2], diesel [Bromberg3] and hard-to-reform biofuels [Cohn] into hydrogen rich gas ($H_2 + CO$). Recent developments in compact plasmatron reformer design at MIT have resulted in substantial decreases in the value of the electrical power requirements. These new developments also increase the lifetime of the electrodes. The development of these devices has been sponsored by the DoE Office of Heavy Vehicle Technologies for the purpose of reducing engine exhaust pollutants by providing hydrogen rich gas for combustion in spark ignition and diesel engines. Compact plasmatron fuel converter technology could also be used with NO_x absorber/adsorber catalysts and particulate traps. NO_x absorber catalysts and particulate traps have been suggested for the decreasing emissions from diesel and spark ignition engines. In order to regenerate the catalyst, a reducing agent is introduced into the exhaust, either directly injected, or in the case of spark ignition engines by running the engine fuel rich during the regenerating period. Use of compact plasmatron reformers for generating hydrogen rich gas for regenerating the catalyst could provide important advantages. This paper describes compact plasmatron fuel converter-aftertreatment catalyst systems that could provide new opportunities for improved diesel engine emissions control.

II. Hydrogen Production from Compact Plasmatron Fuel Converter

Plasmatron fuel converters provide electrical discharges in flowing gases of hydrocarbon fuels and air (and/or other oxidants). The resulting generation of reactive species in the flowing gases along with increased mixing accelerates reformation of hydrocarbon fuels into hydrogen rich gas. Plasmatron fuel converters may also be utilized for increasing enthalpy, further accelerating the reaction rates. These conditions facilitate the reforming of a wide range of hydrocarbon fuels into hydrogen-rich gas without the requirement of using a reformer catalyst. It is possible to eliminate problems associated with reformer catalyst use, such as narrow operating temperature, sensitivity to fuel composition, poisoning, and response time limitations.

By increasing the reaction rates, plasmatron fuel converters can significantly reduce size requirements for effective reforming, increase speed of response and substantially increase fuel flexibility. A wide range of fuels can be converted to hydrogen rich gas, including diesel fuel. The boost provided by the plasma can ensure conditions for partial oxidation reactions to occur with negligible soot production and with a high conversion of hydrocarbon fuel into hydrogen-rich gas.

A plasmatron reformer can be made very small because of reaction accelerating characteristics. Moreover, the capability to rapidly vary plasmatron parameters (energy input, flow rate, product gas composition, etc) make this technology very suitable for application to the dynamic demands for hydrogen-rich gas production in vehicles. It should be possible to produce hydrogen-rich gas for injection into a catalyst in a practically instantaneous amount of time.

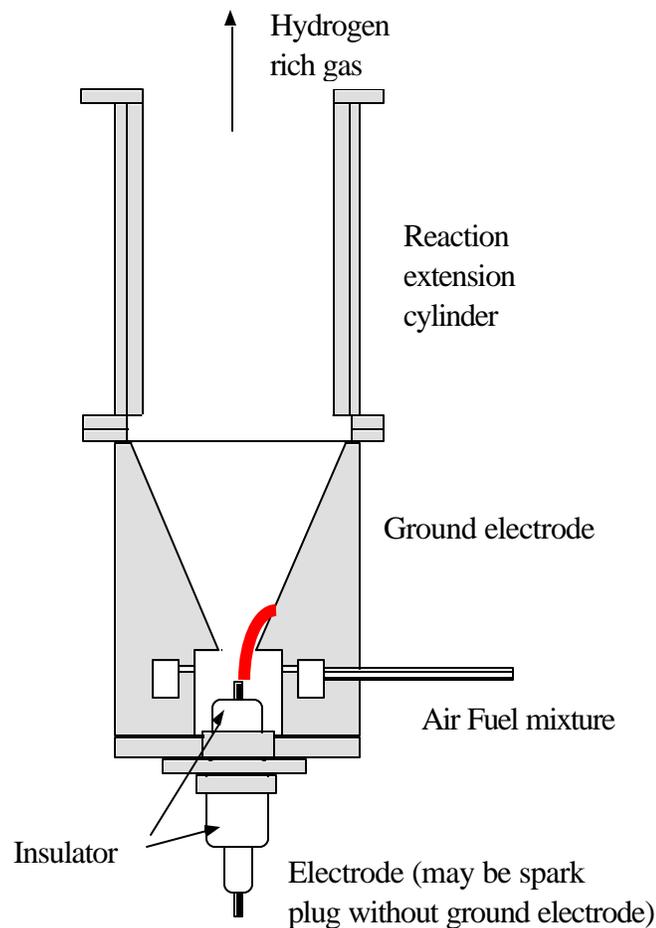


Figure 1. Plasmatron fuel converter device.

Figure 1 shows a diagram of a low current plasmatron reformer. A discharge is established across an electrode gap. Air and fuel are injected in the plasma region. The device operates at atmospheric pressure, with air as the plasma forming gas. Use of a special discharge mode

allows for operation at relatively small electrical powers, at much reduced current relative to earlier compact arc plasmatron fuel reformers developed at MIT. When operating DC, the cathode can be a spark plug. The ground electrode of the spark plug would have been removed. The anode can be a steel or copper cylinder. Neither electrode is water cooled.

Table I shows the typical plasmatron parameters for a low current, low power compact plasmatron fuel converter.

The plasma source is followed by a reaction extension cylinder. A simple heat exchanger can be used downstream from the reaction extension cylinder, both to cool the reformat and to preheat the incoming air and/or fuel.

Table 1.
Parameters of present low current compact plasmatron fuel converter

Power	50-500 W.
Voltage	500-20000 V
Current	15-120 mA
Flow rates H ₂	30-50 liters/min
Height	20 cm
Volume	2 liters

The required electrical energy input to the compact plasmatron fuel converter is on the order of 2% of the heating value of the fuel.

The low current device showed no evidence of soot, even after extended operation. Typical reforming efficiency in partial oxidation mode of operation was around 70% (ratio of heating value of the hydrogen rich gas to heating value of the fuel). It is projected that reforming efficiencies of 80% will be attainable with device/process improvements. Diesel fuel has been successfully reformed using a high current, low voltage compact plasmatron reformer using an arc. Initial tests with the low current compact plasmatron indicate it should also be able to practically reform diesel. However, further testing must be carried out in order to definitely establish this capability.

Compact plasmatron fuel converters have substantial dynamic range. The lower power is determined by the maximum voltage capability of the power supply (the voltage increases with decreasing current), while the highest power is determined by erosion of the electrodes. It is expected that a dynamic range of a factor of 10 is possible without substantial modification to the plasmatron device. This is sufficient to provide the required change in throughput for conventional engines.

Projected parameters for higher throughput versions of low current compact plasmatron fuel converter systems are an H₂ flow rate of 500 liters per minute, and electrical power requirements of 500-1000 W.

III. Exhaust Aftertreatment with Absorber Catalysts

An absorber catalyst can be used for trapping the NO_x, and then chemically reducing NO_x in the system during regeneration periods. The process of trapping could be either catalytic absorption or adsorption, or conventional absorption or adsorption [Campbell]. The catalyst for NO_x treatment is very sensitive to poisoning by sulfur. In order to solve this problem, a sulfur trap is under development [Parks], in conjunction with the development of NO_x absorber catalysts that are more sulfur tolerant. In this case, the sulfur and the NO_x traps are regenerated simultaneously

Experimental studies of combined SO_x and NO_x traps have been carried out recently by Goal Line Technologies [Parks, Parks1]. These studies examined both the use of diesel fuel and hydrogen as reducing agents for trap regeneration. Greater than 90% NO_x removal was obtained. Further work is needed to determine effects of sulfur on catalyst lifetime and the difference between the use of diesel fuel and hydrogen rich gas for catalyst regeneration.

Goal Line Environmental Technologies has recently claimed a 98.9% reduction in NO_x and Hydrocarbon (HC) emissions from diesel engines at their test facility last spring. Using their proprietary catalyst system, attached to a Cummins 50-kW/86-bhp diesel engine, NO_x and HC have been reduced to 0.4 grams per brake horsepower-hour (g/bhph), an emissions reduction that is a full order of magnitude lower than the U.S. EPA's 2002 emission reduction requirement of 4.0 g/bhph for diesel engines.

A similar process may be used for trapping/regenerating particulates. The harmful or noxious emission can be trapped in one system, and then released and treated in a downstream unit (engine or another catalyst).

A recent patent by Daimler Benz [Boegner] describes the regeneration of one of the catalyst while the other is in the adsorbing mode. This patent discusses a motor vehicle exhaust emission control system that has tandem adsorber catalysts connected in parallel for alternate adsorption and desorption. NO_x from the engine goes to the operating adsorber catalyst while the other adsorber catalyst is in the desorption mode. An oxidizing converter is located upstream of the adsorber part and near the engine for oxidation of the NO contained in the exhaust to form NO₂, so as to permit an increase in the NO_x adsorption rate for the nitrogen adsorber parts. A reducing agent is used to regenerate the catalysts.

IV. Advantages of Compact Plasmatron Fuel Converters

The use of compact plasmatron fuel converters in systems for regeneration of these catalysts could provide a number of significant advantages. Hydrogen rich gas is a significantly stronger reducing agent than diesel or gasoline. Use of hydrogen rich gas could potentially provide:

- Reduced poisoning of catalyst (since diesel fuel is not used for regeneration)
- Reduced adverse effects of sulfur (hydrogen-sulfur interactions could reduce sulfur poisoning effects)
- Reduced emissions of hydrocarbons, since it is easier to prevent release of hydrogen and CO than hydrocarbons due to faster catalytic removal
- Higher regeneration effectiveness
- Reduced loss in overall fuel efficiency due to lower requirements on amount of reducing gas and greater ease of reuse of hydrogen rich gas in the engine.
- Shorter regeneration time due to greater reducing capability of hydrogen rich gas and high concentration of reducing gases.

Compact plasmatron reformers are very well suited for the required hydrogen-rich gas generation due to:

- Robustness when operating with diesel fuel
- No soot production
- Very rapid reformer response (< 1 second)
- High efficiency
- Low cost
- Small size and weight
- Warm up capability provided by hot gas, since the hydrogen rich gas from the compact plasmatron fuel converter is hot; catalyst warm-up time could be decreased, or the catalyst could be further heated up during regeneration.

V. Catalyst regeneration by direct injection of hydrogen rich gas using tandem absorber/adsorber catalysts

Figure 2 shows a schematic diagram of a compact plasmatron fuel converter feeding hydrogen rich gas to a NO_x-absorber/adsorber and/or a particulate trap catalyst. In the case of the NO_x absorber catalyst, the injection of hydrogen rich gas reduces the nitrogen in NO and NO₂ to N₂. Hydrogen rich gas (approximately 20% H₂, 20% CO with the balance mainly N₂ and CO₂) is a powerful reducing agent.

In catalyst regeneration by direct injection of hydrogen rich gas, the exhaust flow from the engine is diverted from the catalyst to be regenerated. The diverted exhaust flow may be sent to a second catalyst. Hydrogen rich gas is then rapidly injected onto the catalyst. Some of the gas from close-cycle operation of the NO_x-absorber catalyst needs to be released, in order to prevent pressure buildup in the closed system. The gas from the catalyst during regeneration can be sent to the engine, a second catalyst and/or the plasmatron fuel converter.

There are several variations in which the compact plasmatron fuel converter can be combined with the catalysts. In one variation, the compact plasmatron fuel converter is integrated with the NO_x-absorber catalyst. If there are two NO_x-absorber catalyst units, then two compact plasmatron fuel converters are required. An alternative approach consists of a single plasma fuel converter, directing the hydrogen rich gas into the unit that is being regenerated. This method of operation saves on the cost of the plasma fuel converter units, but requires a high temperature valve. This embodiment is shown in Figure 5. A high temperature EGR valve could be used for this purpose. If the gas from the NO_x-absorber catalyst being regenerated is to be recycled, either in the engine, in the compact plasmatron fuel converter or in the other NO_x-absorber catalyst unit, then it is needed to have a valve at the exhaust to control the flow of the gases.

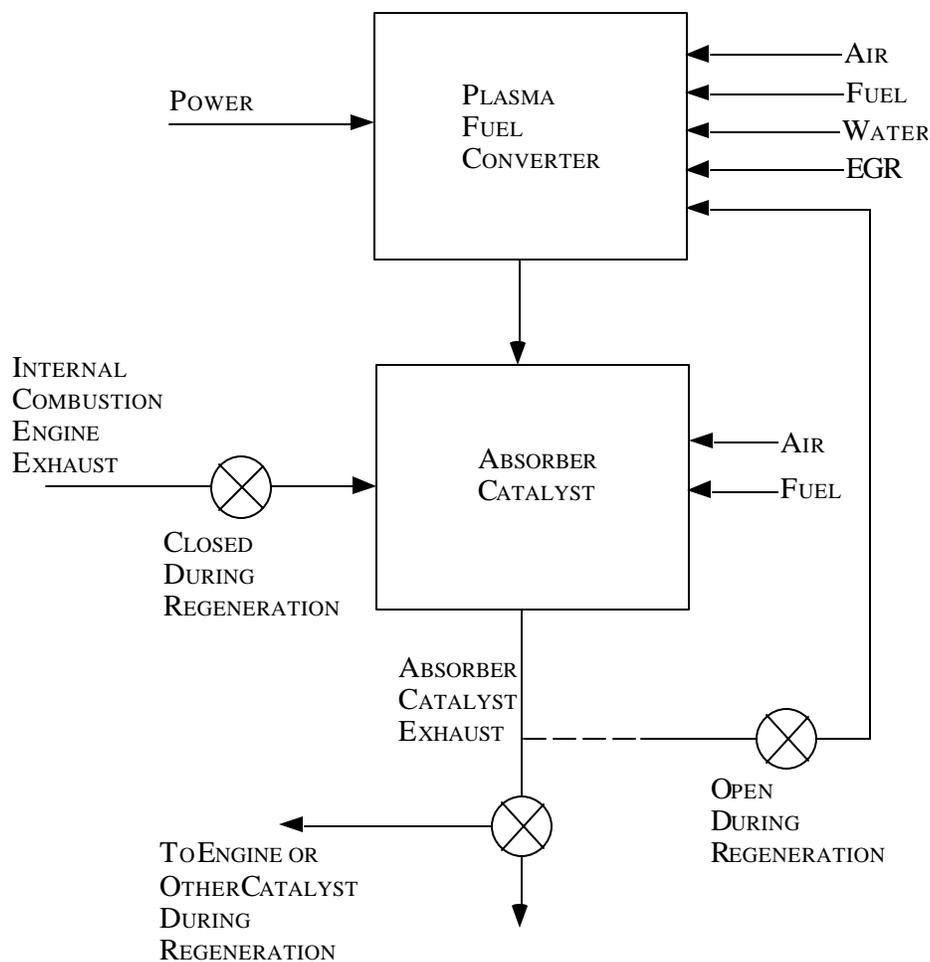


Figure 2. Schematic diagram of a compact plasmatron fuel converter/absorber catalyst system for the treatment of engine exhaust emissions.

The plasma fuel converter may be set to operate continuously while the engine is operational, with hydrogen rich gas normally used in the engine to reduce emissions. During absorber

catalyst regeneration, part or all of the hydrogen rich gas from the plasma fuel converter can be redirected into the absorber catalyst unit. The hydrogen rich gas production rate by the compact plasmatron reformer could also be increased.

The devices described above could also be used, with a modification of the absorber catalyst, to treat other emissions. It may be useful to employ a system that uses a particulate trap that is regenerated by the injection of hydrogen rich gas, with or without an oxidant. In this embodiment, the hydrogen rich gas could also be used, along with some free oxygen, for burning the particulates deposited in the particulate trap. Or if the temperature of the trap is high enough, it could be used for gasification of the particulate, without the use of an oxidant. The absorbing and or gasification processes could be either homogeneous or catalytic in nature.

Illustrative parameters for a compact plasmatron-fuel converter system regenerating an absorber catalyst are shown in Table 2. It is assumed that the hydrogen throughput requirements are twice stoichiometric, and that the CO that is produced in the compact plasmatron fuel converter does not help reducing the catalyst. Hence, there is a margin of a factor of 4 in the flow rate of reducing agent relative to the 'best' possible conditions. The actual amount needs to be experimentally determined. Based on the illustrative parameters in Table 2, if all of the chemical energy in the fuel used for regeneration were lost, the loss in chemical energy would be around 2% of the mechanical energy produced by the engine. Taking engine efficiency into consideration, this loss then translates into less than 1% loss relative to total fuel chemical energy to produce the mechanical power.

Table 2
Compact plasmatron fuel converter/absorber system
Illustrative parameters

Engine NOx production	g/bhp-hr	6		
Engine power	hp (kW)	30 (23)	60 (45)	120 (90)
Engine NOx rate	g/hr	180	360	720
Low current plasmatron				
Average electrical power requirement	W	9	18	35
Plasmatron duty cycle	%	4%	8%	16%
Plasmatron peak power	W	220	220	220
Average chemical power of diesel fuel to plasmatron	kW	0.44	0.88	1.76

VI. Conclusion

NOx and particulate absorber/adsorber catalysts are promising technologies for controlling emissions from diesel engines. Prospects for successful use may be significantly enhanced by systems which combine these technologies with compact plasmatron fuel converters. The requirements on the compact plasmatron fuel converter have been evaluated using conservative assumptions on the regeneration process, and it has been determined that the requirements for the compact plasmatron fuel converter are modest. Short regeneration times, with low duty

cycle, may well be attainable. Although the paper focuses on applications involving absorber/adsorber catalysts in diesel engines, the use of a compact plasmatron fuel converter for production of hydrogen rich gas for regeneration may have other applications. It could also be used for regeneration of SCR catalysts of engines that run near stoichiometric. Compact plasmatron fuel converter-catalyst aftertreatment technologies could play an important role in the effort to meet the urgent need to reduce pollution from diesel vehicles.

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