NO_X REDUCTION FOR LEAN EXHAUST USING PLASMA ASSISTED CATALYSIS

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Introduction

Currently CARB estimates on road diesel vehicles contribute 50% of the NO_X and 78% of the particulates being discharged from mobile sources. Diesel emissions obviously must be reduced if future air quality targets are to be met. A critical technological barrier exists because there are no commercial technologies available, which can reduce NO_X from diesel (lean), exhaust containing 5-15% O_2 concentration.

One promising approach to reducing NO_X and particulates from diesel exhaust is to use a combination of plasma with catalyst. Plasma can be generated thermally or non-thermally.

Thermal plasma is formed by heating the system to an exceedingly high temperature (>2000 ^oC). High temperature requirements for plasma makes thermal plasma inefficient and requires skilful thermal management and hence is considered impractical for mobile applications. Non-thermal plasma directs electrical energy into the creation of free electrons, which in turn react with gaseous species thus creating plasma. A combination of non-thermal plasma with catalysts can be referred to Plasma Assisted Catalysts or PAC.
PAC technology has been demonstrated in stationary sources where non-thermal plasma catalysis is carried out in presence of NH_3 as a reductant. In stationary applications NO is oxidized to HNO_3 and then into ammonium nitrate where it is condensed and removed. This approach is impractical for mobile application because of the ammonia requirement and the ultimate mechanism by which NO_X is removed. However, if a suitable catalyst can be found which can use onboard fuel as reductant then the technology holds a considerable promise.

In addition to the development of an effective catalyst, a non-thermal plasma reactor needs be scaled and demonstrated along with a reliable and cost effective plasma power source and onboard HC source needs to be proven.

Under the work sponsored by DOE and SCAQMD Noxtech is developing a cost effective and reliable PAC system for mobile applications. The goal of the program is to develop a suitable catalyst with the ability to remove high levels of NOx at reasonable space velocities. This new catalyst will then be used to scale the technology to treat exhaust from 80Hp engine and eventually to demonstrate the technology on 200 and 400 Hp engine applications.

Using the 2004 EPA proposed regulation as a standard, it is clear in order for PAC system to be commercially viable it needs to remove NO_x by 70% or better. It is further assumed from past experience that 30,000 HR^{-1} space velocities are necessary to ensure a good compact design.

Accomplishments

Listed below is a summary of major accomplishments achieved so far in the program.

1. 80% NOx reduction demonstrated on a bench scale.

2. Built and tested a prototype solid-state plasma power supply.

3. PAC system capable of treating exhaust from 80 HP Cummins engine installed and operational.

4. Currently 65% NOx reduction from 80 HP engine exhaust is being attained.

Technical Approach

A bench scale system was developed consisting of a plasma reactor operating in series with the catalyst bed. The power to the plasma was supplied via a Thyratron based power supply and Propene as a source of HC was injected upstream of the plasma rector. The exhaust gas was either diesel exhaust or simulated gas containing N_2, O_2, H_2O, NO, NO_2, CO, CO_2, C_3H_6 and SO_2 as a sulfur source.

Figure 1 shows the test cell setup used for gathering data from PAC system. The PAC system is equipped with online analyzers to measure NO, NO_2, NO_X, CO, HC (C_1) and with a 400MHZ Tektronix oscilloscope to determine power deposition into the plasma reactor. Additionally the test cell is equipped with a Nicolet 550 FTIR and a Gas Chromatograph for additional gas analysis. The test cell is also equipped to measure system pressure drop and the exhaust gas flow rate. The bench scale system also included temperature controllers to control exhaust gas temperature for the plasma and catalyst bed.

Using the laboratory setup more than 27 potential catalysts were tested and the best material found to date is an A9 catalyst which has demonstrated more than 80% NO_X reduction capability (see figure 2). A9 further demonstrated an ability to operate over a broad temperature range (250-450 ^oC) and is relatively sulfur tolerant (see figure 3). The A9 catalyst when combined with a CZ1 catalyst formed no undesirable by-products when tested with FTIR (see table1).

Having demonstrated A9 catalyst activity preliminary endurance testing was conducted using simulated gases and diesel exhaust (see figure 4).

Using the A9 catalyst a system was scaled up usi-ng the design and operating criteria developed during bench scale studies (see figure 5).

A PAC system being tested to treating exhaust from an 80 Hp Cummins Engine is shown in figure 6. Preliminary data indicates 65% NO_X reduction is being consistently achieved at steady state operation and pressure drop across the system is only 25" of H_2O.

One of the limitations of the present system is the plasma power source. The plasma power source currently being used is Thyratron based, which has a limited life and is not cost effective. As a result Noxtech is building a cost effective, reliable, and very efficient solid-state power supply. The solid-state prototype power supply currently being tested at Noxtech measures only 12"X12"X3" and is capable of providing up to 300 Watts (see figure 7). The solid-state power supply was built from off the shelf components. Estimated size based on the prototype for a power supply to treat exhaust gas from an 80 Hp engine is only 12"X12"X10" or less than 1 FT^3.

Future Work

Future work regarding the PAC system will focus on scaling up the system to treat exhaust from 200 Hp and 400 Hp diesel engines. Catalyst investigation will continue to have a signifycant priority. The solid-state power supply will be scaled up to treat the exhaust from 80, 200, and 400 HP engines in a step up scaling manner. Future work will also concentrate on integrating on-board HC source to remove NO_X and particulates.

In conclusion a suitable catalyst capable of removing 70% NO_x has been developed. The system is built, installed, and is being tested to treat exhaust from an 80 Hp Cummins engine. Preliminary results indicate a 65% NO_x reduction is being achieved at steady state.

Figure 1 Test cell setup for Data Collection

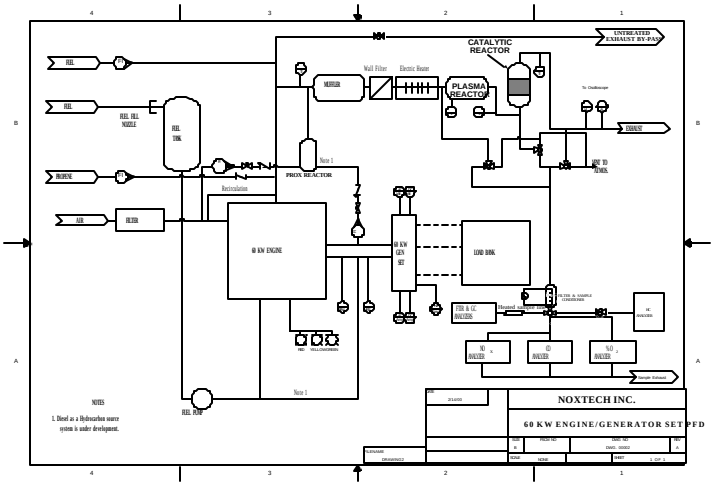


Figure 2 A9 Catalyst Performance at various Temperatures

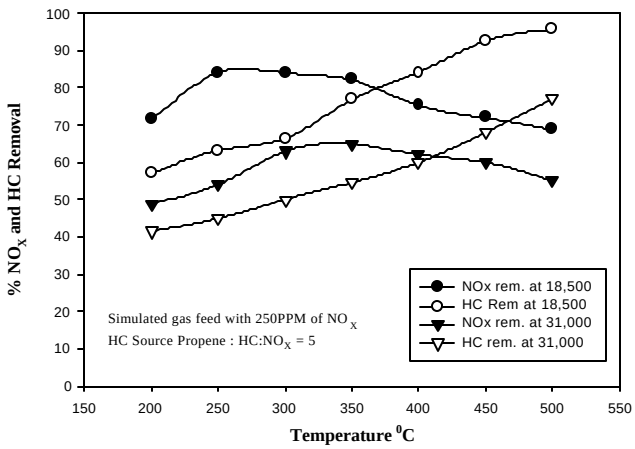
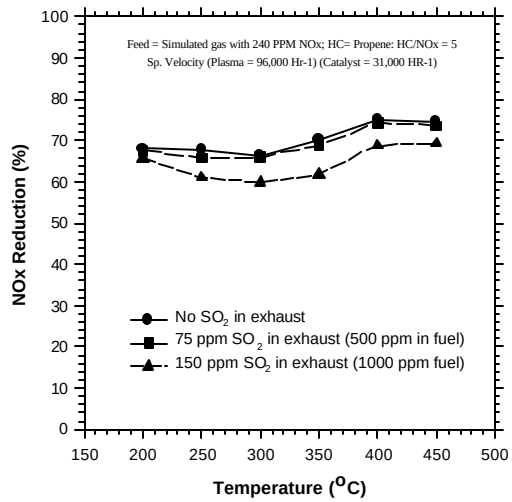


Figure 3 Effect of Sulfur on A9 Catalyst performance



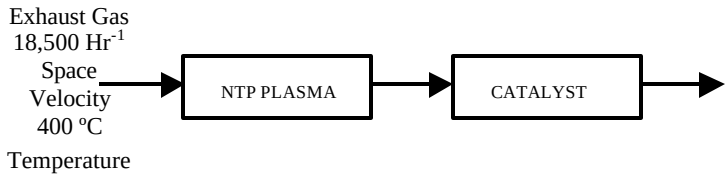


Figure 4 Endurance testing of A9 Catalyst

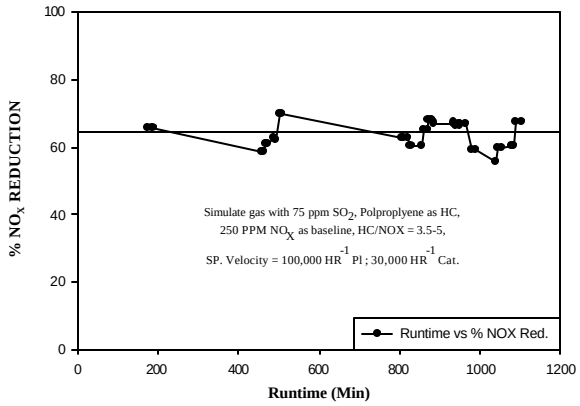


Table 1:By-Product Formation using FTIR analysis

Species (PPM)	IN Feed Gas	After Plasma	After Catalyst
NO	240	4	83
NO2	10	224	30
N2O	1300	720	252
Propene (C1)	8	238	94
Formaldehyde	2	130	11
Acetaldehyde	10	126	3
Nitrous Acid	0	6	0
Nitric Acid	0	0	0
Methyl Nitrate	0	0	0
Ozone	0	10	0
Methanol	0	37	0
Ethanol	30	0	0
Sulfur Dioxide	37	36	15
TOTAL N (Bal)	**250**	**251**	**139**

Figure 5 MECHANICAL DESIGN OF PAC SYSTEM

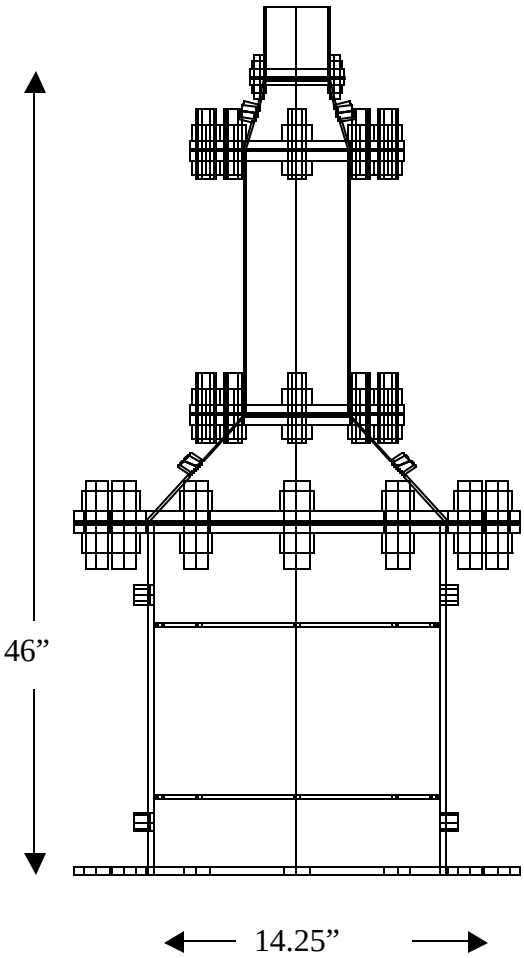
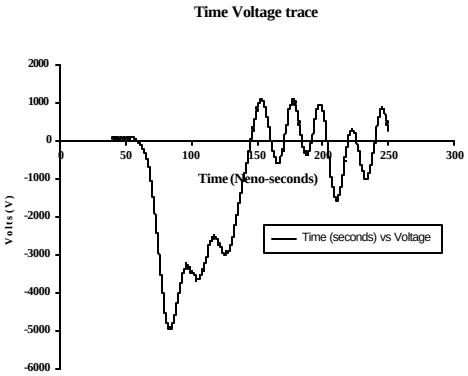


Figure 6 80 HP PAC SYSTEM UNDERGOING TESTING



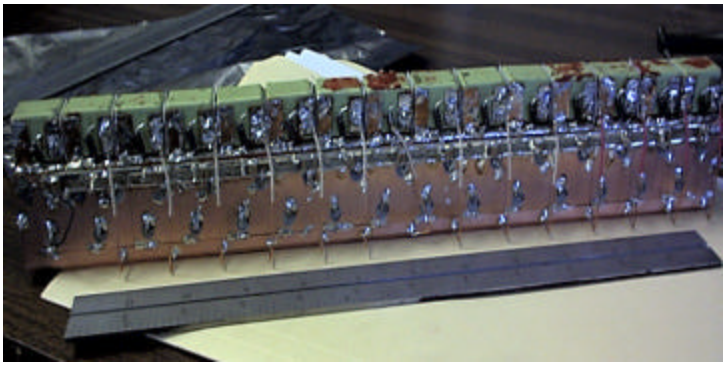


Figure 7 Solid State Power prototype power supply with Oscilloscope trace