Demonstration of Catalytic NOx Reduction System Using Trace Hydrogen Injection

Grant ICAT 00-2

MEI-ARB01-FR

Final Report

Reporting Period:
February 20, 2001 – April 1 2004

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1.0 Executive Summary

1.1 Project Summary

Makel Engineering Inc. (MEI) conducted pilot demonstration program under a grant from the California Air Resources Board (ARB) to develop a practical, low cost NOx emissions reduction system Suitable for lean burn natural gas fuel stationary power generators. This program directly supported the objectives of the ARB’s Comprehensive Work Plan. In particular, this project supported the strategy in the 1994 State Implementation Plan (SIP) for achieving lower NOx emissions. The overall objectives of this project where:

- Build a prototype NOx emissions reduction system
- Perform design optimization tests
- Complete a field test demonstration

The resulting NOx emissions reduction system consists of a hydrogen production source, injection system, catalytic converter, sensors and control system. Laboratory tests were conducted at MEI and University of California at Berkeley (UCB). Once the design was completed optimization testing was conducted, and system refinements where made and the laboratory scale hardware was scaled up to be suitable for use on 100 to 250 kW class engines. Finally, a field test demonstration was commenced and performance data was gathered on a 100kw genset running in the field.

The resulting product addresses the need for effective NOx reduction systems for lean burn combustion devices. The initial target market for this technology is lean burn, natural gas powered reciprocating engines (stationary power generators). However, this technology may ultimately be adapted to diesel power generators and potentially to heavy-duty vehicles (CIDI engines) in conjunction with new particulate control systems and/or clean burning fuels currently under development.

In the presence of excess oxygen, conventional catalytic converters have very low NOx conversion rates. To address this limitation of existing technology, MEI’s research represents an effective NOx reduction systems for lean burn combustion devices. The MicroNOx Trace Hydrogen Injection NOx reduction system is an exhaust after treatment system that is intended to be a system that can be used with existing natural gas or propane fueled lean burn engines without any modification to the engine control system. This system is applicable to both the retrofit and OEM engine/gen-set market. The system consists of a set of catalytic converters that are in parallel with each other and used in a cyclic fashion. The converters consist of NOx storage catalyst stages upstream of conventional 3-way catalytic converter. The converters are used to store NOx and are then regenerated with a hydrogen/carbon monoxide product stream from an external microchannel based reformer. System control is achieved using a NOx sensor downstream of the converters. With proper sizing of the NOx storage catalysts, the required fuel flow for regeneration of the catalyst is less than 0.5% of the total fuel flow to the engine.

This research culminated in a field-test of a 100 kW natural gas generator configured with a MicroNOx exhaust system. Figure 1 shows the genset and MicroNOx exhaust system. Specifications for the system is provided in Table 1

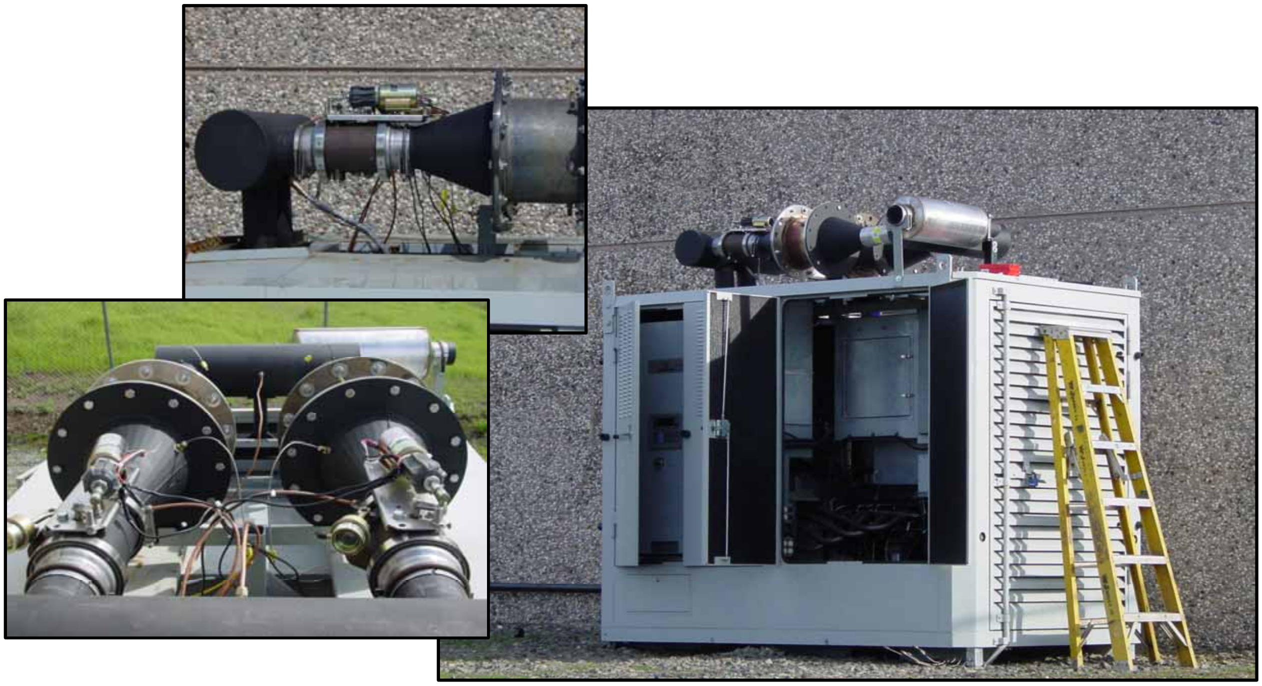


Figure 1. 100 kW Natural Gas Generator Set and MicroNOx Exhaust System For Field Testing

Table 1. MicroNOx System Operating Specifications

Fuel Type	Natural Gas or Propane (1)
Exhaust Inlet Temperature	350 C min
Maximum Inlet Temperature	TBD (estimated less than 550 C)
NOx Reduction	Greater than 80%
Fuel Consumption	Approximately 0.5%
Inlet Power	24 VDC
Power Consumption	12 W

The field test site was the Butte Community College water pumping system site demonstrates the technology's ability to utilize trace hydrogen to improve the NOx reduction efficiency of catalytic clean up of NOx emissions. The Butte College water pumping station relies on an aerobic digester system and a series of evaporation ponds. The field test generator replaces the 175kw diesel generator and operated offline through a switching system. The BCC waste treatment plant is based on a digester system. Raw sewage is pumped into a pumping well and run through two 5 hp sewage grinders. Two 7.5 hp lift pumps (Figure 2) feed the sewage into an aerobic digester (Figure 3). Two 25 hp blower (Figure 4) aerate the sewage in the digester system. These blowers and pumps run continuously alternating between electric motors. The processed sewage is released into a gravity flow system, pouring into a series of holding ponds where it evaporates.

Figure 2. Lifter Pumps.

Figure 3. Aerobic Digester.

Figure 4. 25 Hp blowers and existing diesel gen set

Testing with the final version of the system developed on this program demonstrated a NOx reduction of over 80% with up to 5% oxygen in the engine exhaust. The system was successfully operated over a nine-month field test with numerous starts as the system was typically used for sporadic short periods (typically a few hours).

2.0 System Design

The project was broken down into three major tasks.

Task 1. NOx Reduction System Design and Fabrication
In task 1 the prototype NOx reduction system was designed and fabricated. The nominal size was sufficient to supply trace hydrogen to an engine or gas turbine sized for light commercial and residential neighborhood use (approximately 100 kW).

Task 2. Testing of Prototype Systems
Task 2 was devoted to the testing of the NOx reduction system on operating engines. Tests were conducted at UC Berkeley using gas turbine and reciprocating engines. The prototype system design was optimized and sensor feedback systems were tested.

Task 3. Field Trial of Prototype System
Task 3 combined a H_2 generator and catalyst at the Butte Community College water treatment facility in Northern California. A nine-month field trial was conducted and the data was compared to conventional instrumentation.

2.1 Theory of Operation

The basic operating principle of the catalytic systems employing NOx storage and reduction have been described in the open literature (Theis, 2002) (Rodrigues 2001). While exact mechanisms of NOx storage under oxidizing exhaust conditions and that of nitrate desorption/reduction under reducing exhaust conditions are complex, a simplified model is shown in Figure 5. The NO in the exhaust stream is oxidized with the help of oxygen on the platinum component of the NOx storage catalyst. The resulting NO_2 migrates to the NOx storage material (barium carbonate) and is reversibly stored there as a nitrate. Once the NOx storage capacity of the barium carbonate is exhausted, the gas environment is changed from oxygen rich to hydrogen and carbon monoxide rich. Under these conditions NO and oxygen are released from the barium nitrate surface and can be subsequently reduced to N2 with a conventional three-way catalytic converter and exhausted. Practical design consideration dictate that the storage catalyst be capable of storage times exceeding 30 seconds and that regeneration time be accomplished in no more time than the storage. It is desirable to use the least amount of fuel to create the fuel rich conditions used during regeneration (i.e. release of stored NOx) and conversion on the three way converter.

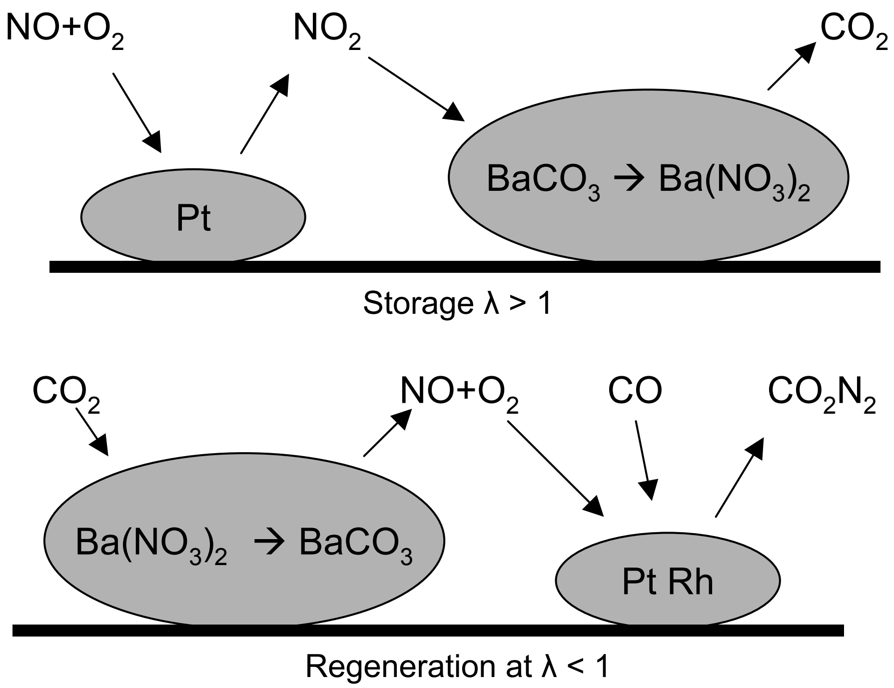


Figure 5 NOx Reduction Mechanism With Regeneration.

2.2 NOx Reduction System Overview

The NOx reduction system consists of two parallel exhaust treatment lines, which are alternate between exhaust treatment and catalyst regeneration. Thermocouples allow monitoring temperature of the exhaust gas. An oxygen sensor provides useful data when adjusting the flow/air ratio for the engine. The concentration of NOx is monitored using a NOx analyzer. A switch valve allows the selection of samples upstream or downstream of the exhaust treatment. Figure 6 and Figure 7 illustrate how the flow paths are switched between the two parallel exhaust treatment lines.

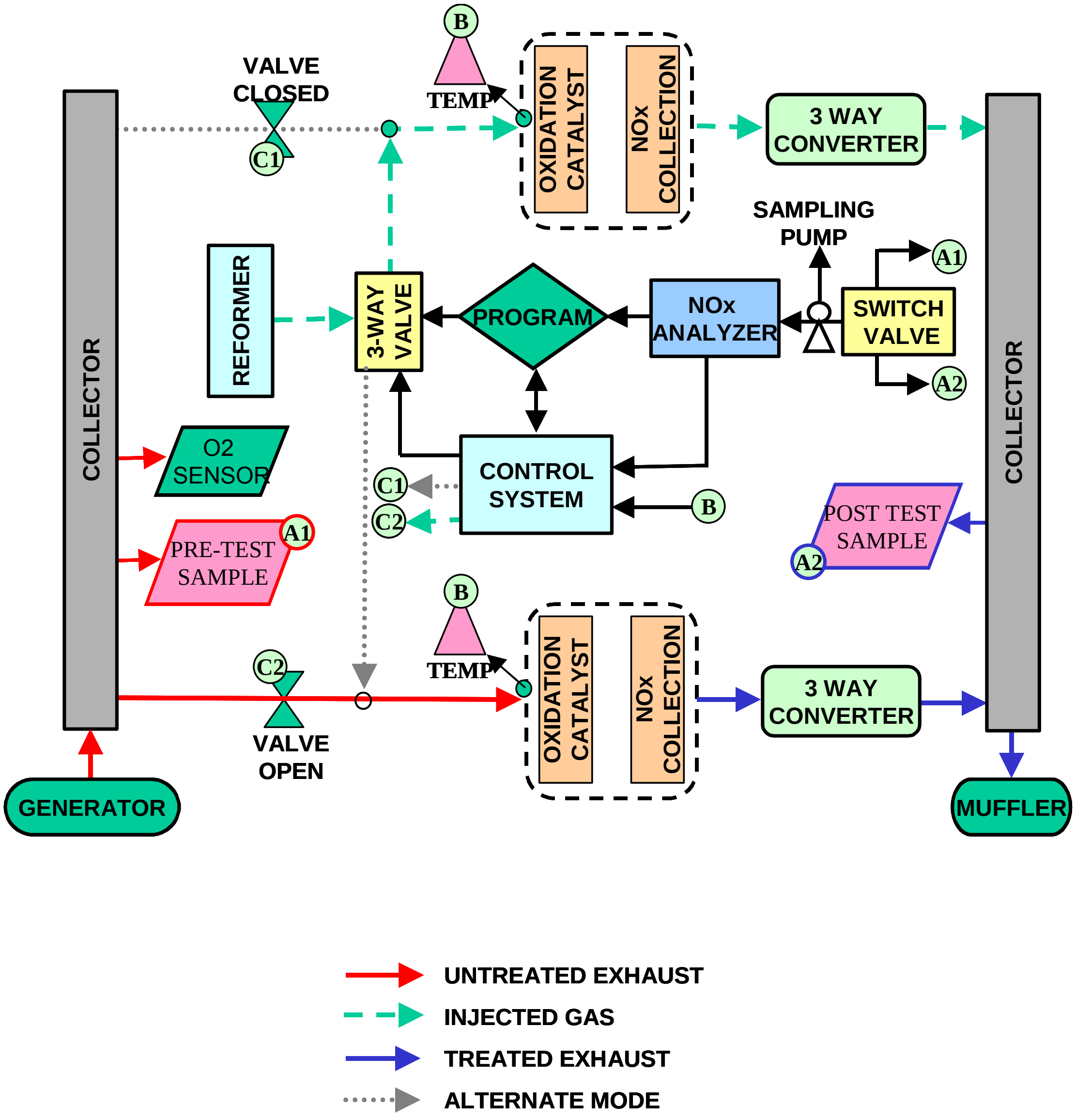


Figure 6. Schematic of NOx Reduction System Switching Regenerator/Exhaust Treatment - Flow Through Bottom Section, While Regeneration of Top Section

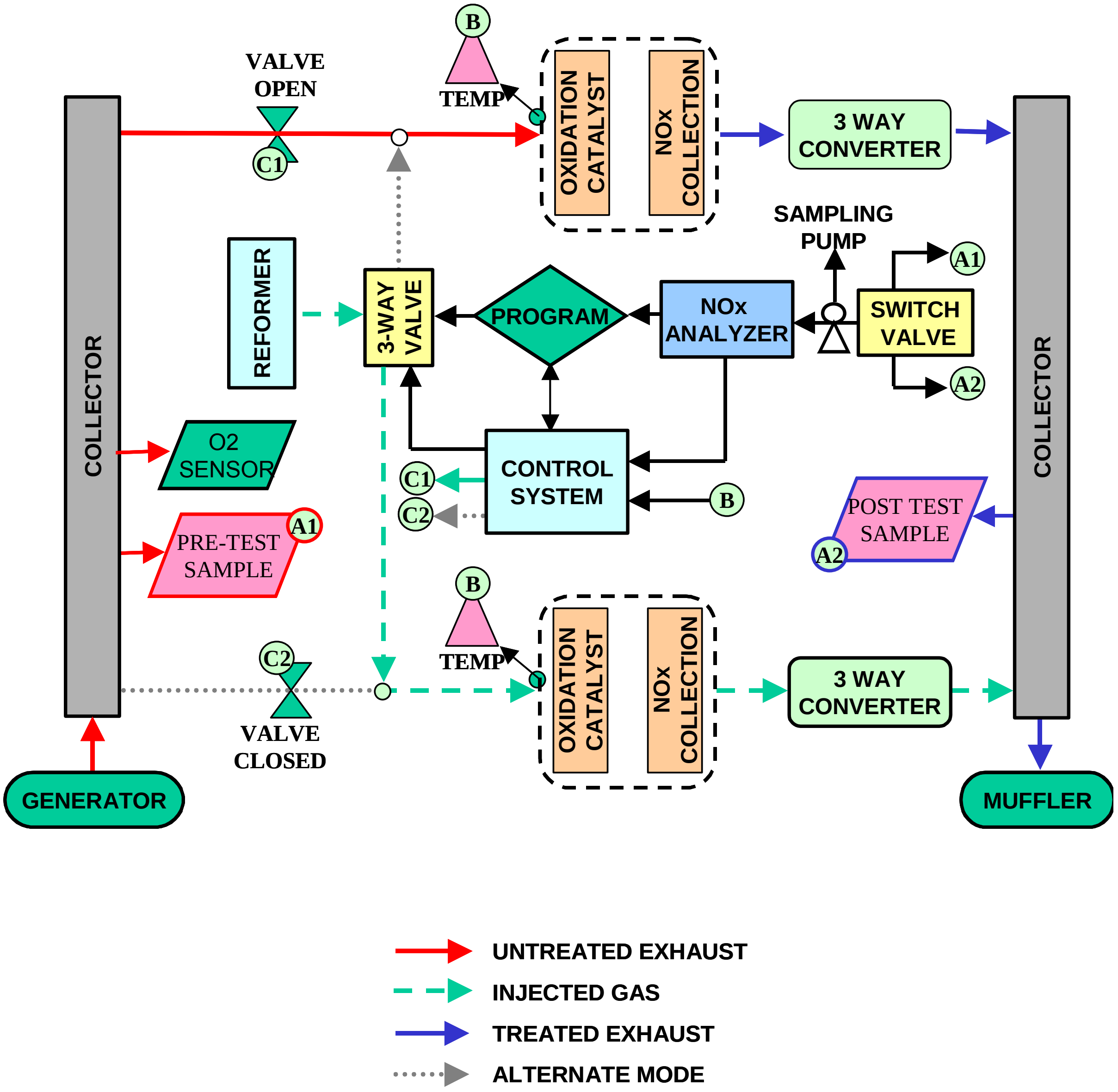


Figure 7. Schematic of NOx Reduction System Switching Regenerator/Exhaust Treatment - Flow Through Top Section, While Regeneration of Bottom Section

The line running in exhaust treatment mode directs the engine exhaust through the sections of catalyst beds. As shown in Figure 6, solenoid valve C2 is open. There are a total of three sections: the oxidation catalyst, the NOx collection and the conventional 3-way converter. The oxidation catalyst and the NOx collection are the key components to enable NOx reduction.

The line running in regeneration mode blocks the engine exhaust (close solenoid valve C1) and directs the regenerating stream through the sections of catalyst beds (using the 3-way solenoid valve). The regenerating stream is a mix of hydrogen and carbon monoxide. This mixture can be supplied from gas bottles (during scoping experiments) or from a methane reformer.

The reformer converts methane and air into hydrogen and carbon monoxide. To provide stable flow, the reformer gas supplies to the reformer use sonic orifices. With sufficient gas inlet pressure, the flow is independent on the pressure variations downstream of the orifice. The reformer is equipped with a heater and closed loop temperature control. However, the reformation of methane is an exothermic reaction, once it gets initiated. A possible modification of the system is to take advantage of the exhaust heat to initiate the reaction, eliminating the need of a heater.

2.3 System Prototype Design

The prototype system developed was intended to be a versitile design that would allow several modes of operation to be evaluated and would allow components such as catalysts to be easily removed for evaluation and upgraded as necessary. The two primary modes of operation were the cyclic storage and reduction described in section 2.2 and the a pulsed or continuous flow mode such as that demonstrated by Toyota . When the prototype system was modified for use in field testing the pulsed mode features of the system were abandoned based on results indicating the a lower fuel consumption penalty with the cyclic (alternate regeneration) mode of operation. These modes for the prototype are summarized below.

2.3.1 Alternate Regeneration and NOx Reduction

While one converter is used to reduce NOx, the other is regenerated with H_2. Periodically, each converter switches between NOx reduction and regeneration with H_2. Adjustment of the duration of each cycle is controlled by the NOx sensor response: if the NOx levels start to rise, the cycle is shortened. Likewise, the hydrogen flow can also be adjusted to ensure the converter is regenerated within the duration of the regeneration cycle, but not too high to cause waste.

2.3.2 Pulsed Hydrogen Injection

Only the catalytic converter equipped with the injector is used (the untreated exhaust and the hydrogen are directed to this converter). Hydrogen is injected using 1/16 in tubes placed axially on a distribution manifold in a spoke wheel configuration. Each 1/16 in tube has 0.005 in orifices to distribute hydrogen through the catalyst bed. Figure 8 shows the converter equipped with the injector assembly and the detail of the hydrogen delivery section. The fuel injector assembly enables pulsed hydrogen injection. Supplying hydrogen in pulsed flow, instead of continuous flow, causes momentary high concentrations of hydrogen, making the catalyst regeneration more effective. This technique should minimize the amount of hydrogen required to reduce NOx emissions to the desired levels. Figure 9 the results of flow tests performed in the hydrogen injector. The tests indicate that large flow rates can be achieved with small inlet pressure.

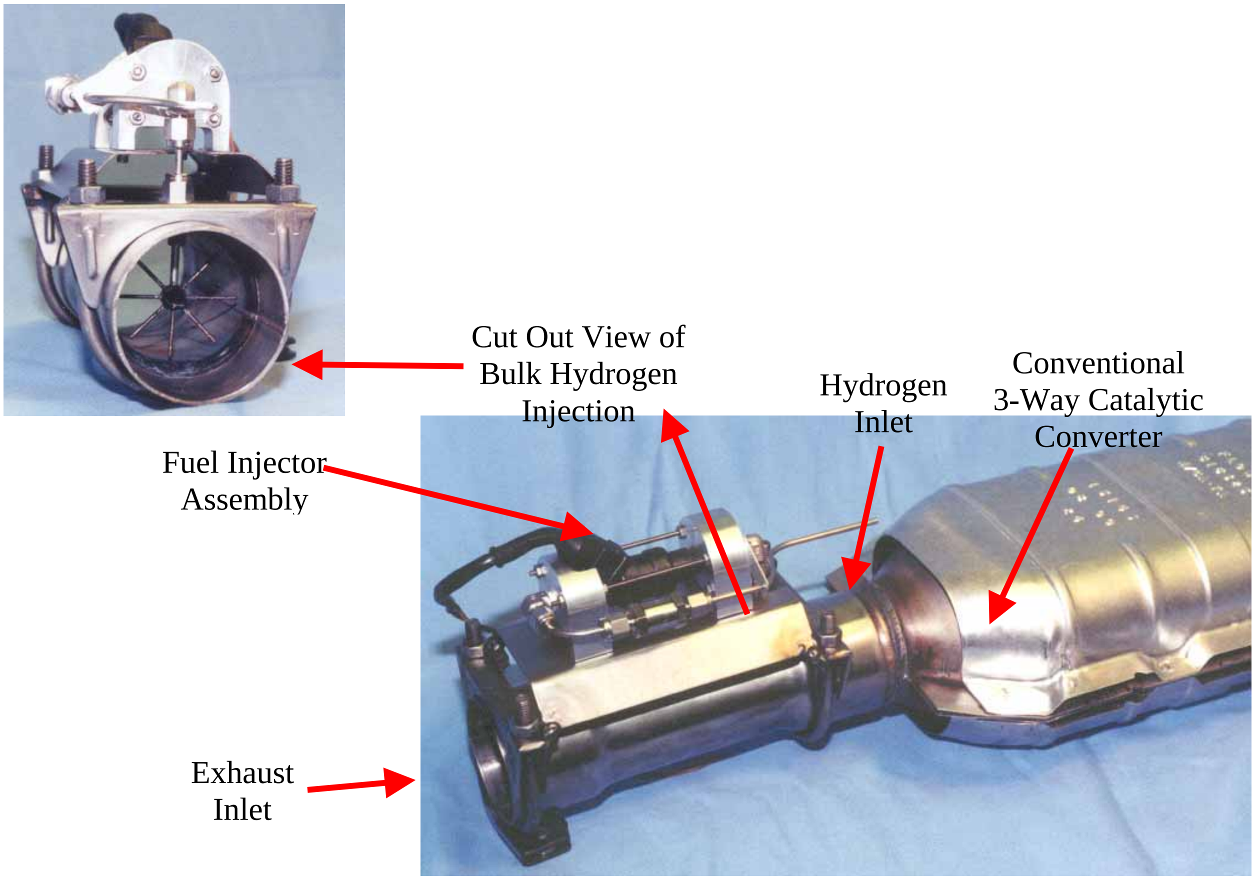


Figure 8. Hydrogen Injector Coupled with Three Way Catalytic Converter

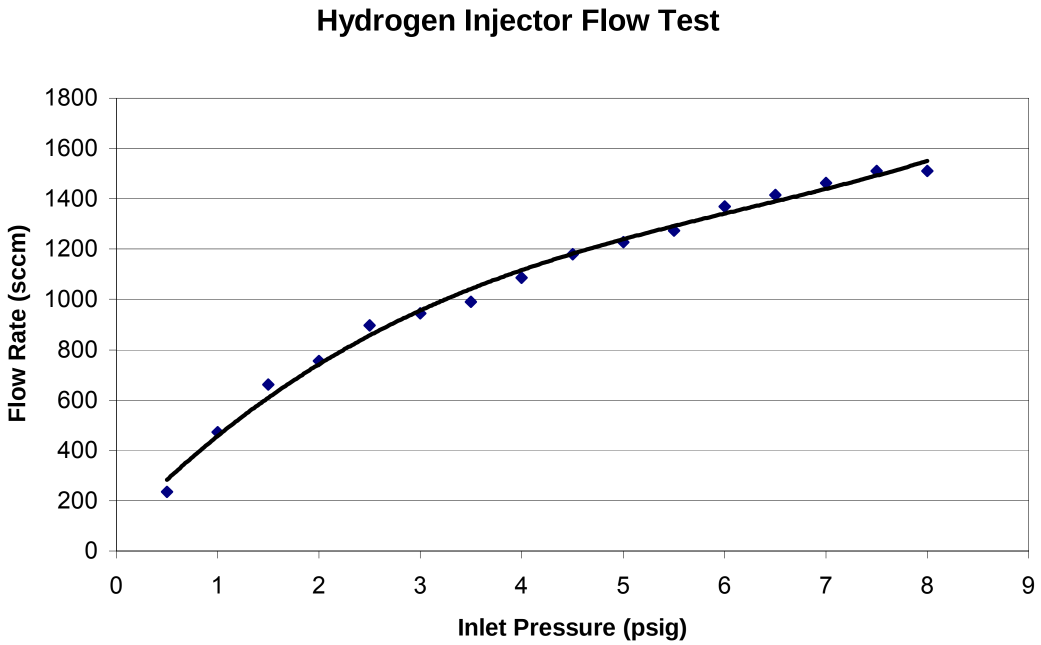


Figure 9. Injector Flow Test Using Nitrogen as a Test Gas – Effect of Inlet Pressure on the Flow Throughput

2.3.3 NOx Sensor

The NOx reduction system has a NOx sensor downstream of the combined exits from both catalytic converters, as shown in Figure 10. The feedback from the NOx sensor enables regulation of the cycles between regeneration and NOx reduction when operating in Mode 1, or the pulsed injection of hydrogen, when operating in Mode 2.

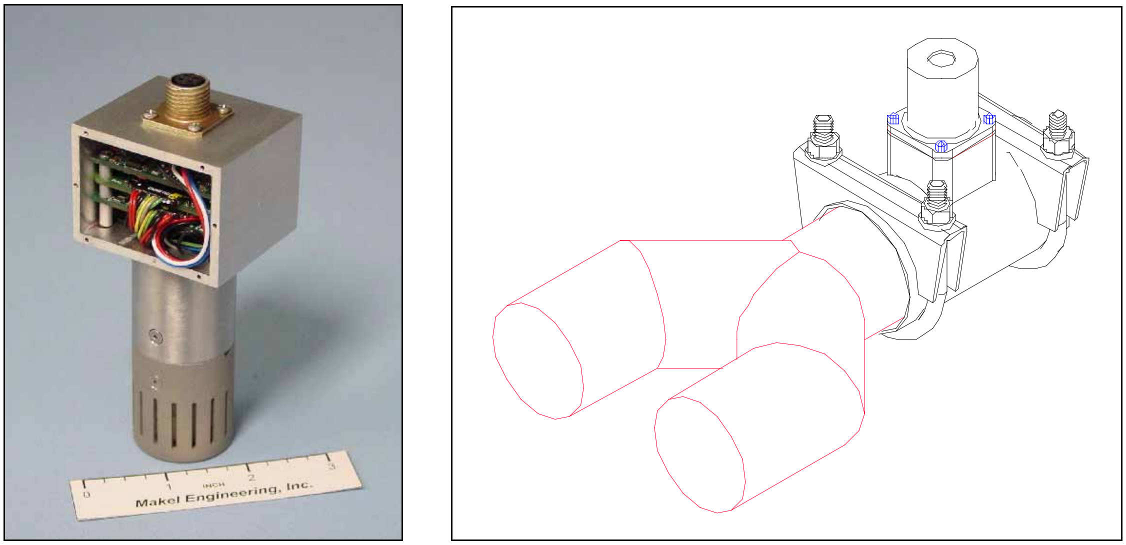
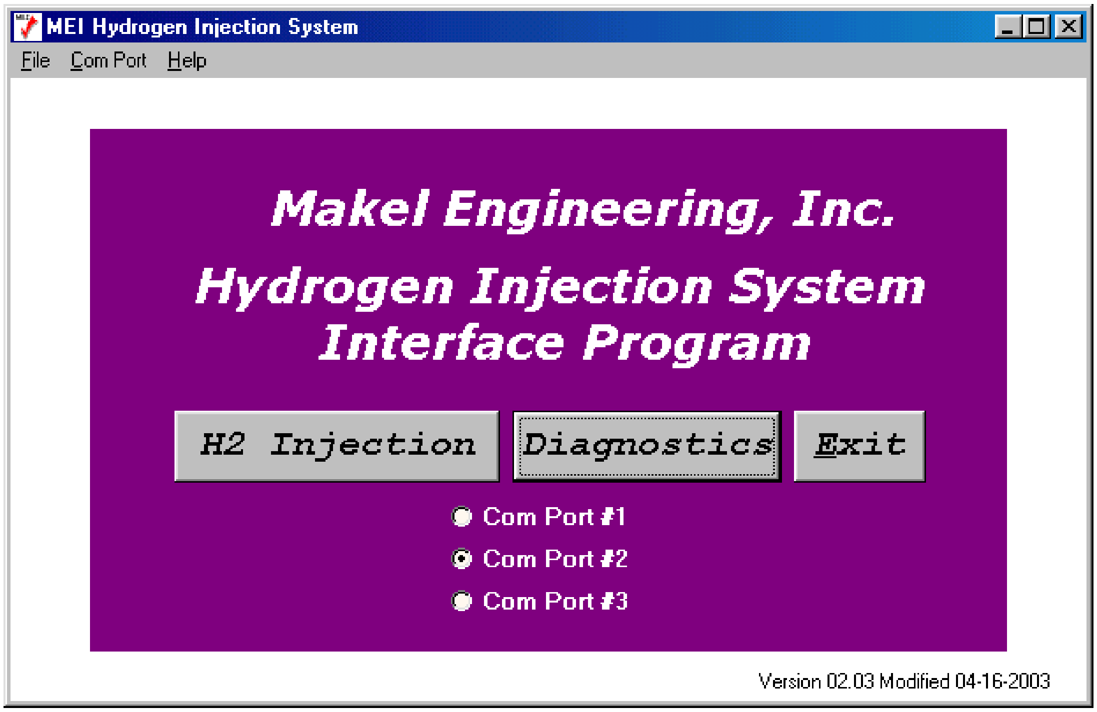


Figure 10. NOx Sensor and NOx Sensor Installed at the Combined Exhaust from the Catalytic Converters

2.3.4 Control System

The operation of the NOx reduction system is controlled by a PC computer using a graphical software interface. When the interface is started, Figure 11 allows the user to select communication port and to enter the H_2 reformate injection control. The communication port enables the data logging from optional gas sensing devices. Figure 12 shows the hydrogen injection control screen. This screen is used to control the operation of the NOx reduction system as well as to collect data from the tests.

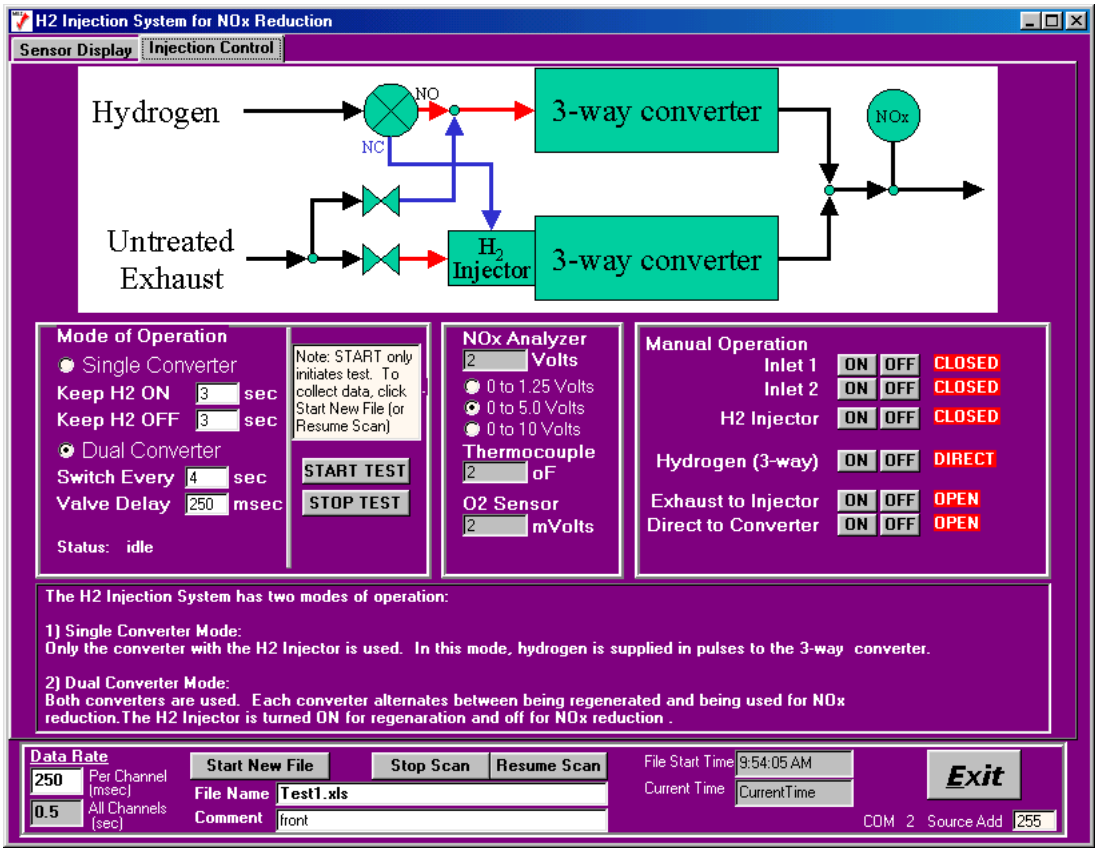
The “Dual Converter” option of the software is used to control the alternating flow between the exhaust treatment side and the regeneration side. The “Single Converter” option is used to control a smaller development system built in the early stages of this program.



○ Com Port #1
● Com Port #2
○ Com Port #3

Version 02.03 Modified 04-16-2003

Figure 11. Control Program Main Screen



Mode of Operation

○ Single Converter

Keep H2 ON 3 sec

Keep H2 OFF 3 sec

● Dual Converter

Switch Every 4 sec

Valve Delay 250 msec

Status: idle

Note: START only initiates test. To collect data, click Start New File (or Resume Scan)

NOx Analyzer

2 Volts

○ 0 to 1.25 Volts

● 0 to 5.0 Volts

○ 0 to 10 Volts

Thermocouple

2 oF

O2 Sensor

2 mVolts

Manual Operation

Inlet 1 ON OFF CLOSED

Inlet 2 ON OFF CLOSED

H2 Injector ON OFF CLOSED

Hydrogen (3-way) ON OFF DIRECT

Exhaust to Injector ON OFF OPEN

Direct to Converter ON OFF OPEN

The H2 Injection System has two modes of operation:

1) Single Converter Mode:
Only the converter with the H2 Injector is used. In this mode, hydrogen is supplied in pulses to the 3-way converter.

2) Dual Converter Mode:
Both converters are used. Each converter alternates between being regenerated and being used for NOx reduction.The H2 Injector is turned ON for regenaration and off for NOx reduction .

Data Rate

250 Per Channel (msec)

0.5 All Channels (sec)

File Name Test1.xls

Comment front

File Start Time 9:54:05 AM

Current Time CurrentTime

COM 2 Source Add 255

Figure 12. NOx Reduction System Control Screen

MEI bases the embebbed control of the NOx reduction system on compact control electronics developed for other products. Two of the CPU boards shown in Figure 13 and an interface board make the complete electronics stack. Figure 14 shows a block diagram of the control electronics and its interfaces.

Figure 13. CPU Board

The control electronics stack consists of the following components and functions:

CPU Boards:

- Closed loop temperature control of the hydrocarbon reformer
- Control of gas inlet valves
- Control of the 3-way valves for the hydrogen injection and untreated exhaust
- Control of hydrogen reformate injection setup
- Data acquisition and serial communications
- Closed loop temperature control of the NOx sensor
- Linearization of NOx sensor signal
- Data acquisition and serial communications

Interface Board:

- Input power conditioning to the CPU boards. This includes transient voltage protection, overvoltage protection, and fuse.
- Transient voltage protection of RS-485 communication lines that connect to the CPU boards.
- Switching field effect transistor for the sensor heater resistor.
- Switching field effect transistor for the H_2 injection relay driver.
- Connection of the NO_X sensor leads (sensor, heater and RTD)
- Connection of the Reformer heater and RTD
- Connection of the valves and relays

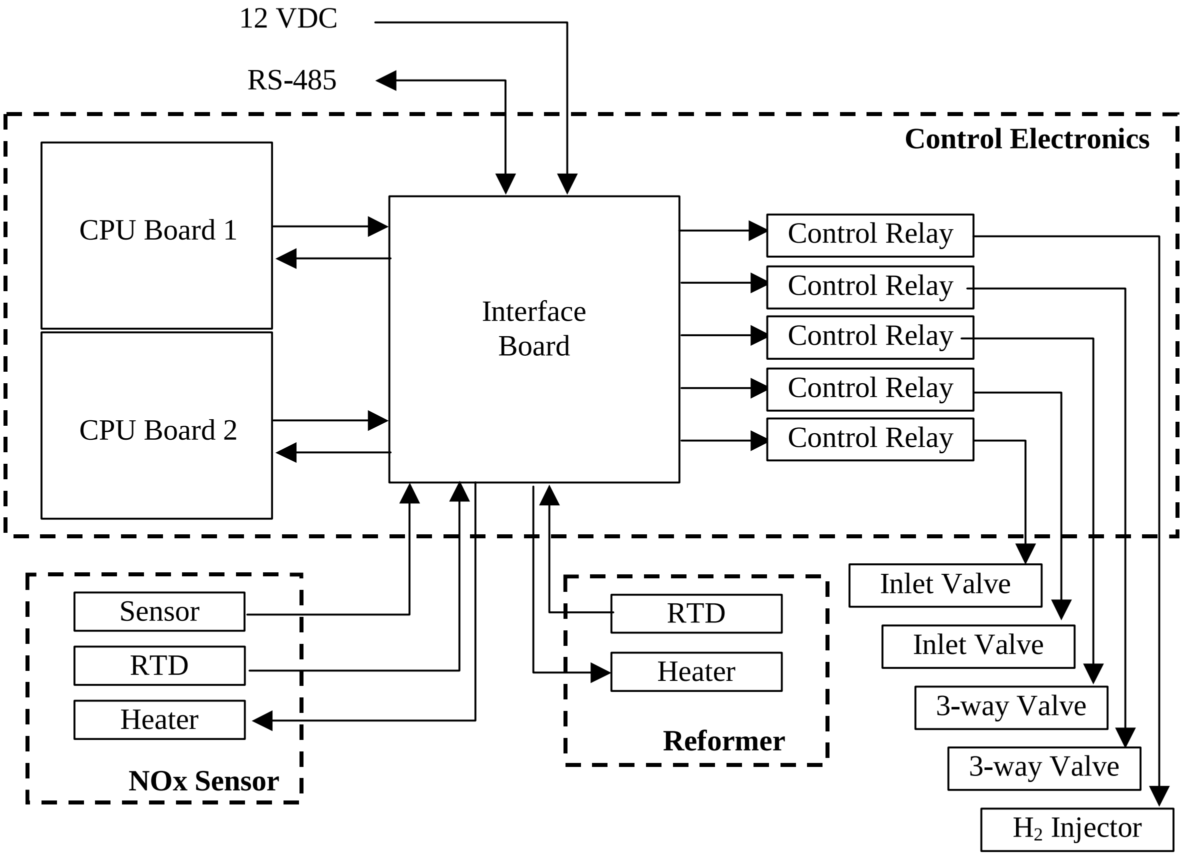


Figure 14. Block Diagram of Control Electronics and Interfaces

2.3.5 Hydrocarbon Reformer

The most complete version of the NOx reduction system incorporates a hydrogen generation system. The hydrogen generation was a microchannel hydrocarbon reformer, which has been developed to generate hydrogen for fuel cells. Since the hydrogen source for the NOx reduction system does not need to be free of carbon dioxide, the reformer consists only of a partial oxidation reactor. The initial target of the NOx reduction system is natural gas engines. The partial oxidation of methane to produce hydrogen occurs according to the following equation:

$$CH_4 + \frac{1}{2} O_2 \rightarrow 2H_2 + CO \qquad \Delta H = -35.6 \text{ kJ/mole}$$

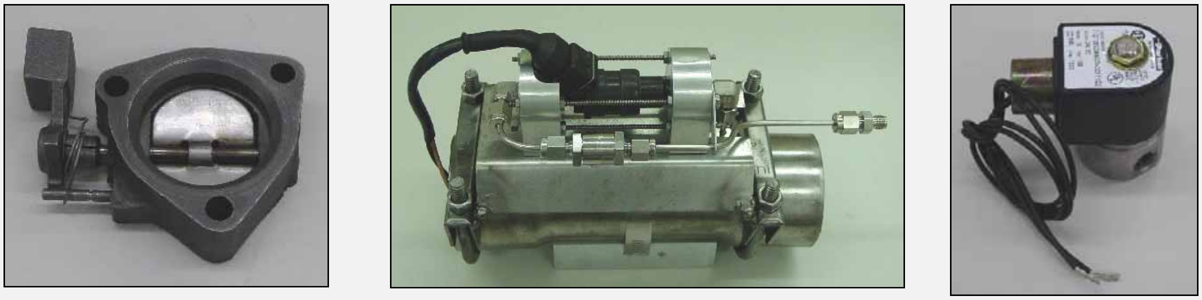
A microchannel reformer was fabricated for integration into the prototype. Figure 15 shows the reformer and the reformer packed in the solid insulation fabricated for mounting on the prototype.

Figure 15. Microchannel Hydrocarbon Reformer with Insulation

2.3.6 Integrated NOx Reduction System

A prototype of an integrated NOx reduction system was fabricated. The system was tested at UC Berkeley test facilities, the system uses two catalytic converters and it was operated in two different modes.

Figure 16 shows some of the components used and Figure 17 shows the prototype fabricated.



Untreated Exhaust Valve Injector Assembly Solenoid Valve

Catalytic Converter

Figure 16. Selected Components Used in Prototype

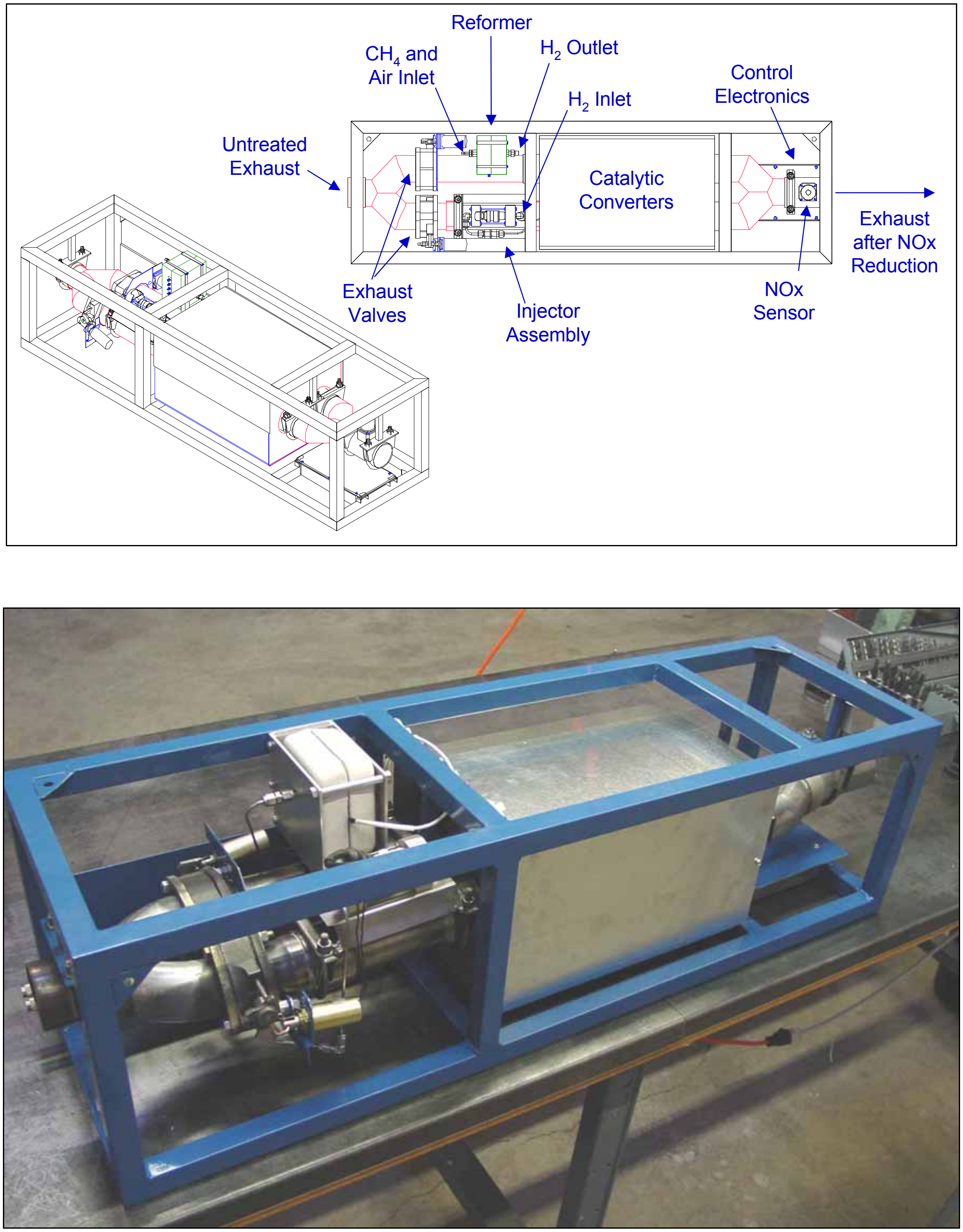


Figure 17. Integrated NOx Reduction System

2.3.7 Catalyst Preparation

NOx oxidation and NOx absorption catalysts were prepared in honeycomb substrates. An image of this honeycomb substrate is pictured below.

The procedure used for the preparation of the catalysts follows:
NOx oxidation catalyst preparation:

1) Alumina washcoat was applied to the honeycomb.
2) Excess coating was removed and substrate was allowed to dry.
3) Substrate was heated in air to high temperatures for firing of the washcoat.
4) A solution of platinum chloride was prepared with proper concentration.
5) After firing the washcoat, platinum was impregnated into the honeycomb substrate by dipping it into the solution.
6) Excess solution was removed and honeycomb substrate was dried in the oven and subsequently fired in air.
7) Substrate was heated under hydrogen atmosphere, resulting in platinum catalyst.

NOx absorption catalyst preparation:

1) Alumina washcoat was applied to the honeycomb.
2) Excess coating was removed and substrate was allowed to dry.
3) Substrate was heated in air to high temperatures for firing of the washcoat.
4) A solution of barium nitrate was prepared with proper concentration.
5) After firing the washcoat, barium nitrate was impregnated into the honeycomb substrate by dipping it into the solution.
6) Excess solution was removed and substrate was dried in the oven.
7) Honeycomb substrate fired under carbon dioxide atmosphere resulting in barium carbonate.

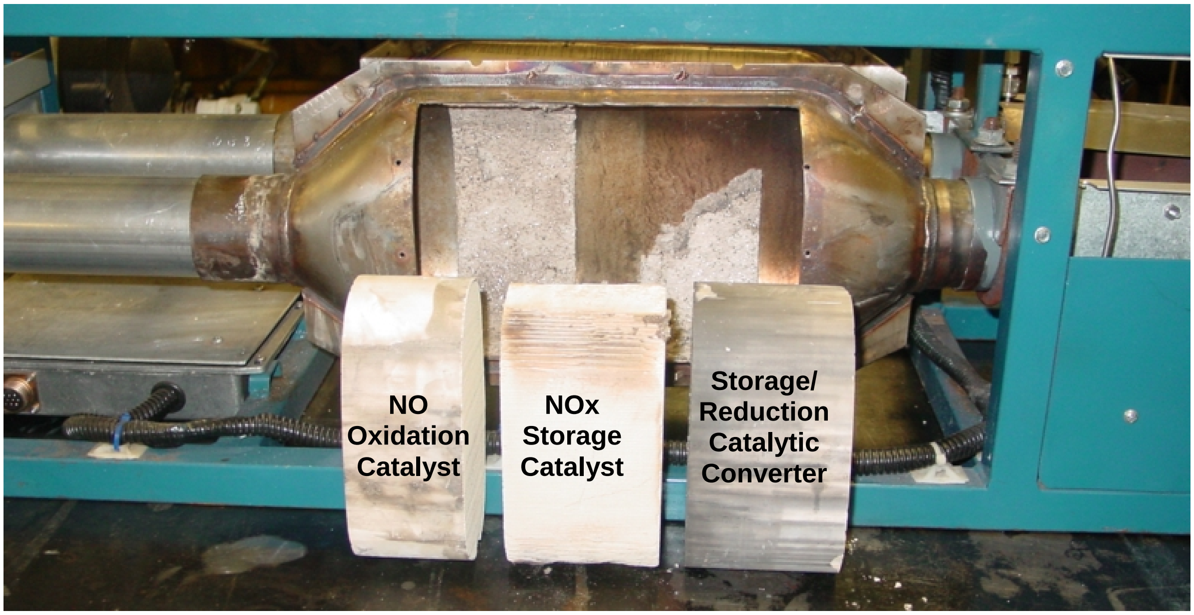


Figure 18. Segmented Catalyst With Oxidation, NOx Storage, and 3-Way Catalyst Sections.

3.0 Prototype System Testing

3.1 Bench Testing of NOx Reduction

Initial tests simulating the dual catalytic converter design were conducted using a single catalytic converter, with time, the high oxygen content seems to cause the regeneration with hydrogen to be less effective. Using the dual catalytic design, when the catalyst is being regenerated, there is no NOx or Oxygen being flown, allowing full regeneration in a shorter period of time, using less hydrogen. The prototype allowed operation in both modes enabling verification of bench test results with engine tests.

Bench test results used a single catalyst bed and alternated the flow of simulated exhaust and the regenerating gases (hydrogen, methane). The simulated exhaust had 170ppm NOx and ~9% O2, in a nitrogen background. Figure 19 shows a comparison of results using 1% hydrogen/carbon monoxide and 5% methane as the reducing gases. Figure 20 illustrates the effect of oxygen concentration on the catalyst regeneration efficiency, using 10% hydrogen/carbon monoxide mixture.

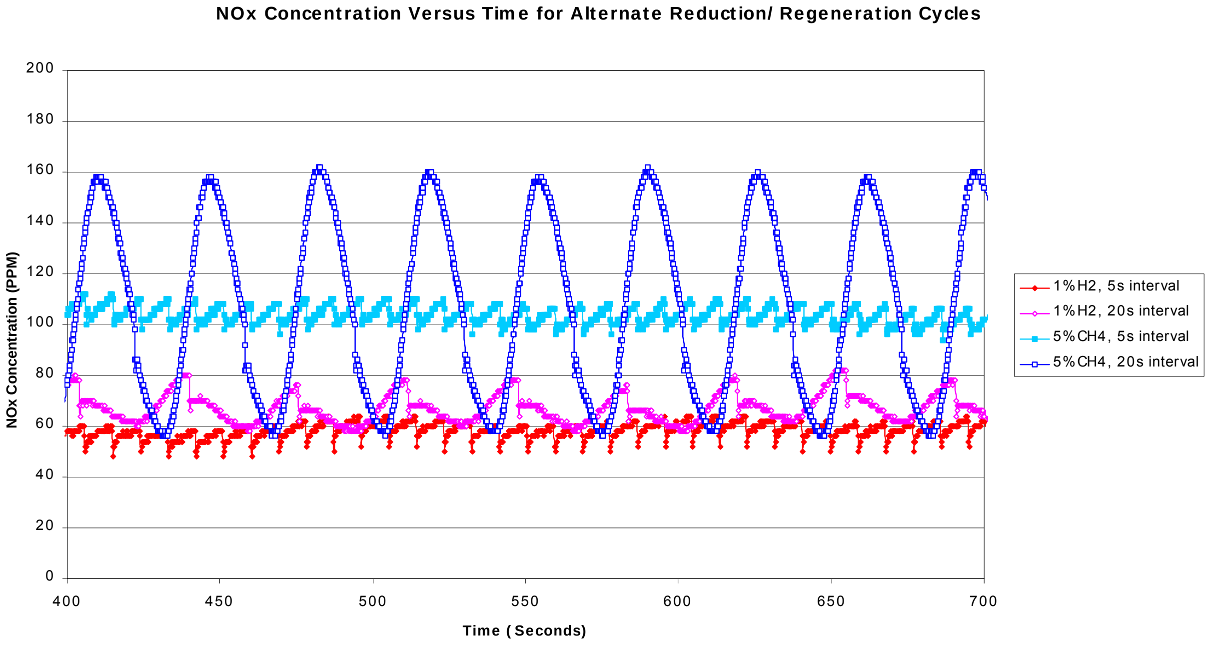


Figure 19. Comparison of Methane and Hydrogen Injection for the Reduction of NOx

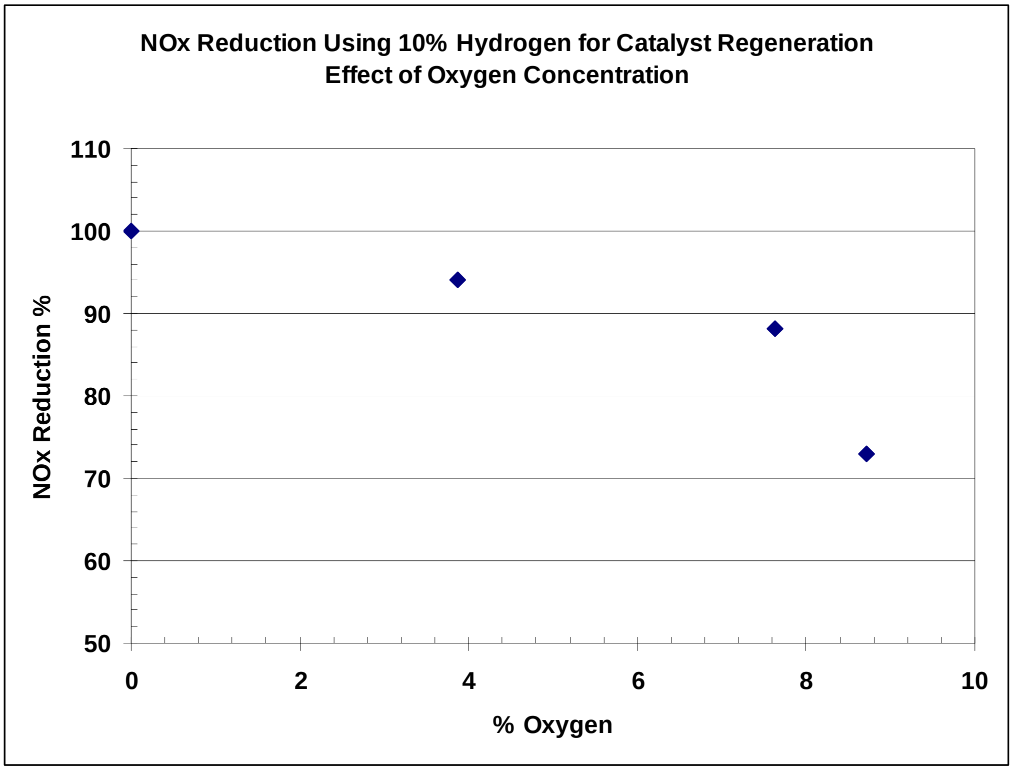


Figure 20. Effect of Oxygen Concentration on NOx Catalyst Regeneration

3.2 Lab Engine Testing of NOx Reduction

Lab engine testing was performed at the UC Berkeley Combustion Laboratory. The NOx Reduction System was hooked up to the exhaust system of a Cummins B5.9 diesel engine (see Figure 21 and Figure 22) operated with ultra low sulfur bio-diesel. The output of the engine ran into a dynamometer to create a variable load for different operating conditions.

A diesel particulate filter between the engine exhaust and the NO_x Reduction System removed soot and other pollutants from the exhaust stream. This filter was a cordierite honeycomb structure measuring 10.5 in x 12 in, with 100 cell per square inch. The particulate filter removes > 95% of all solid particulate matter from the engine and the catalyst coating ($50gm/ft^3$ of platinum) oxidizes gas phase CO and hydrocarbons. Over time, the filter became loaded with particulate matter, increasing the pressure drop across the filter. Operating the filter at 400^oC for 30-60 minutes lead to oxidation of the collected particulate, therefore regenerating it for further use. This filter was sized for engines between about 200 and 400 Hp and it should last 1000's of hours if it does not suffer uncontrolled regeneration.

Figure 21. Prototype NOx Reduction System Installed Downstream of Cummins B5.9

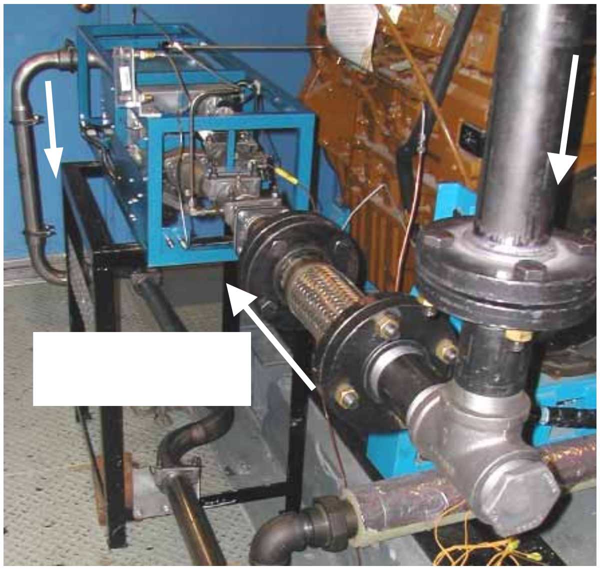


Figure 22. Prototype Close Up

The Cummins B5.9 diesel engine (Figure 23) is a medium-duty, 6-cylinder, 5.9 L, direct injected, turbocharged and aftercooled engine rated at 131 kW (175 bhp) at 2500 rpm. Peak torque is 570 N-m (420 ft-lb) at 1600 rpm. The engine has two valves per cylinder and employs a bowl-in-piston combustion chamber design. It runs at a compression ratio of 17.2 to 1. It has a bore and stroke of 4.02 in (102 mm) and 4.72 in (120 mm) respectively, making the total engine displacement 359 cubic in. (5.9 L). Engine specifications are listed in Table 2

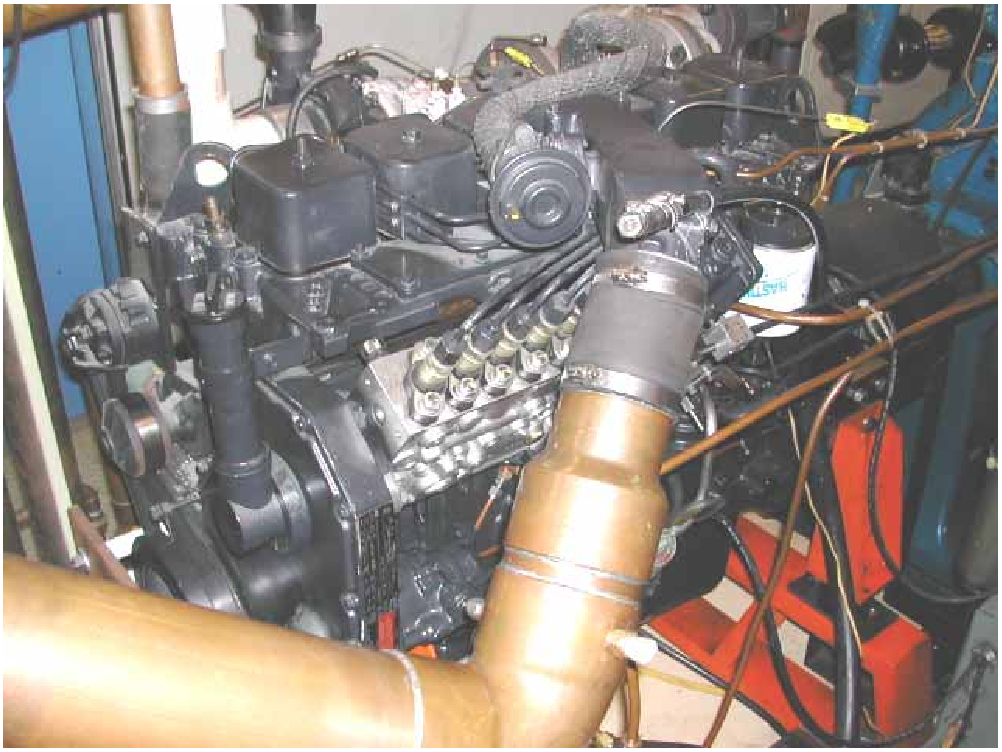


Figure 23. Cummins B5.9

Table 2. Cummins B5.9 Engine Specifications

Date of Manufacture	12-08-1993
Configuration	6 cylinder inline
Displacement	5.88 L (359 in^3)
Horsepower rating	175 bhp @ 2500 rpm
Torque rating	420 ft-lb @ 1600 rpm
Aspiration	turbocharged and aftercooled
Valves	2 per cylinder (12 total)
Bore	102 mm (4.02 in)
Stroke	120 mm (4.72 in)
Compression ratio	17.6:1
Injection timing	11.5° BTDC fixed

Fuel injection on the B5.9 engine was achieved with a Bosch P7100 PE type inline pump, capable of injection pressures of up to 115 MPa. The high injection pressures are comparable to those found in the most modern diesel engines. Injection timing was mechanically controlled and fixed at 11.5º BTC. Although this feature differs from the computer-controlled fuel injection systems that are commonly utilized in newer engines, the injection timing has a less significant effect on PM emissions. It can, however, influence NO_x formation through its effect on peak cylinder temperatures.

If installed in an on-road vehicle, the B5.9 engine would typically be equipped with an air-to-air intercooler that would provide sufficient charge air cooling as the vehicle travels at speed. However, since the engine is mounted on a stationary engine test stand, the intercooler (a Garrett model 485000-1) was placed in a water tank with controllable water flow. A Mid-West type 1519 eddy current dynamometer with a 415 hp capacity provided load to the engine.

3.3 Instrumentation

The engine operation is monitored by several instruments. Figure 24 shows some of the equipment used for monitoring the tests. Output torque is measured by a load cell. Airflow into the engine is measured with a turbine air meter. Engine RPM and fuel consumption can also be monitored.

Thermocouples are installed on the engine at various locations to monitor fluid temperatures. The temperature measurements are as follows:

- Air intake
- Water inlet and outlet
- Engine oil
- Each of the six exhaust ports
- Combined exhaust for cylinders 1-3 and 4-6 going into the turbo
- Exhaust after coming out of the turbo

Gaseous emissions are monitored using a system of Horiba gas analyzers as listed in Table 3. NO_x is determined through chemiluminescence (Horiba CLA-220 Chemiluminescent Analyzer mfg # 570580021). HC is measured with a flame ionization analyzer, and CO and CO_2 is detected based on infrared absorption. The CLA-220 was designed specifically for measuring the NO and NOx concentrations in automotive exhaust in parts per million. Its selectable ranges enable full-scale output from 10 ppm up to 1000 ppm. To measure NO concentration, the analyzer first makes ozone (O_3). Then it reacts the ozone with the exhaust gases. NO and ozone react to produce NO_2 and O_2. 10% of the resulting NO_2 molecules are electrically excited and therefore give of photons. Measuring the level of photons, or chemiluminescence, determines how much NOx is present in the exhaust stream. This measurement is taken with silicon photodiodes, not a photomultiplier tube, so that a wider range of wavelengths can be detected. In order to get total NOx levels, the NO_2 must be converted to NO so it will react with the ozone. This is done by reacting NO_2 with C to produce NO and CO. The total level of NO then reflects the total level of NOx in the exhaust.

Figure 24. UC Berkeley Combustion Lab Instrumentation

Table 3. Equipment for Monitoring of Gaseous Emissions

Species	**Instrument**
NO_x	Horiba Instruments CLA-220 chemiluminescent analyzer
HC	Horiba Instruments FMA-220 flame ionization analyzer
CO, CO_2	Horiba Instruments AIA-220 infrared analyzer

3.4 Prototype Test Results

Initial tests were conducted with the system and the heat loss from the particulate trap and exhaust line was found to be too great resulting in gas inlet temperature to the NOx reduction system of less than 400 F. In addition, the valves that control the direction of the exhaust through the system were found to not fully close under full flow operation. To correct these problems the exhaust system was insulated and the timing on the valves was changed to open the closed valve, before closing the other valve rather than operating the valves simultaneously.

Table 4 shows the best results from diesel engine tests with inlet temperature to the converter of approximately 250 –300 C using the system in mode 1 (alternating injection). The regeneration time was set to 3 seconds for the tests. The reduction of NOx from 40 to 50% with 8 to 12% oxygen in the exhaust is consistent with bench tests results. Trace gas injection with this high of oxygen concentration and the same hydrogen/carbon monoxide flow yielded a reduction of less than 10%.

Table 4. NOx Reduction Results With Hydrogen Reformate Flow of 5 sfc/hr

Sample type	Mode:	Torque:	RPMs:	CO2 %:	HC ppm:	NOx ppm:	NO ppm:	O2 %:	Inlet Temp (F)	% Reduction
pre-catalyzer	4	86.0	1517	5.76%	0.9		285	11.35%	422	
"	4	86.3	1531	5.75%	1.8	511		10.38%	422	
post-catalyzer	4	86.4	1530	5.77%	2.4	533		11.65%	423	
"	4	87.0	1531	5.80%	2.4		312	11.65%	423	
post-catalyzer w/ H2	4	86.4	1526	5.81%	2.4	312		11.58%	427	41%
"	4	86.7	1526	5.81%	2.4		215	11.58%	427	31%
pre-catalyzer	6	137.7	1529	8.40%	1.2	778		8.25%	503	
"	6	137.9	1530	8.34%	0.9		500	8.28%	503	
post-catalyzer	6	138.7	1525	8.50%	6	847		8.15%	506	
"	6	138.6	1524	8.48%	6.3		615	8.15%	506	
post-catalyzer w/ H2	6	138.5	1521	8.51%	7.2	426		8.10%	513	50%
"	6	139.4	1519	8.51%	7.5		302	8.10%	513	51%
pre-catalyzer	9	109.8	2527	6.18%	0.9	278		11.05%	520	
"	9	109.2	2528	6.12%	0.9		149	11.13%	520	
post-catalyzer	9	109.1	2523	6.14%	3.9		196	11.08%	523	
"	9	110.0	2523	6.16%	4.2	300		11.05%	523	
post-catalyzer w/ H2	9	110.0	2520	6.14%	4.5	169		11.08%	526	44%
"	9	110.3	2522	6.15%	4.5		112	11.08%	526	43%

Testing continued with lean exhaust from a natural gas engine where the oxygen operating conditions in the exhaust ranged from 2 to 6%. NOx after treatment is required and with a single stage of regeneration, we achieved 80% reduction in NOx. To achieve 75% to 80% reduction with diesel engine exhaust the system requires two stages of regeneration with approximately 50% reduction in each stage.

Test results with the MicroNOx system operating with the Cummins B6 are shown in Figure 25. The system demonstrated over an approximate 0 to 70% reduction of NOx. The data show that the performance of the system is improved with reduced regeneration cycle times. The best data was obtained with approximately 20% of the exhaust flow. The remaining 80% of the flow was bypassed to the facility exhaust. With higher exhaust flows the space velocity in the catalyst was determined to be too high and less NOx reduction was achieved. With less than 20% of the exhaust flow the catalyst temperature could not be maintained above 500 F.

The NO_2 storage capability of the barium carbonate catalyst stage is shown in Figure 26. The catalyst is capable of storing NO_2, but not NO, so a simple oxidation catalyst is used upstream in the final system or the catalyst is co-impregnated with platinum.

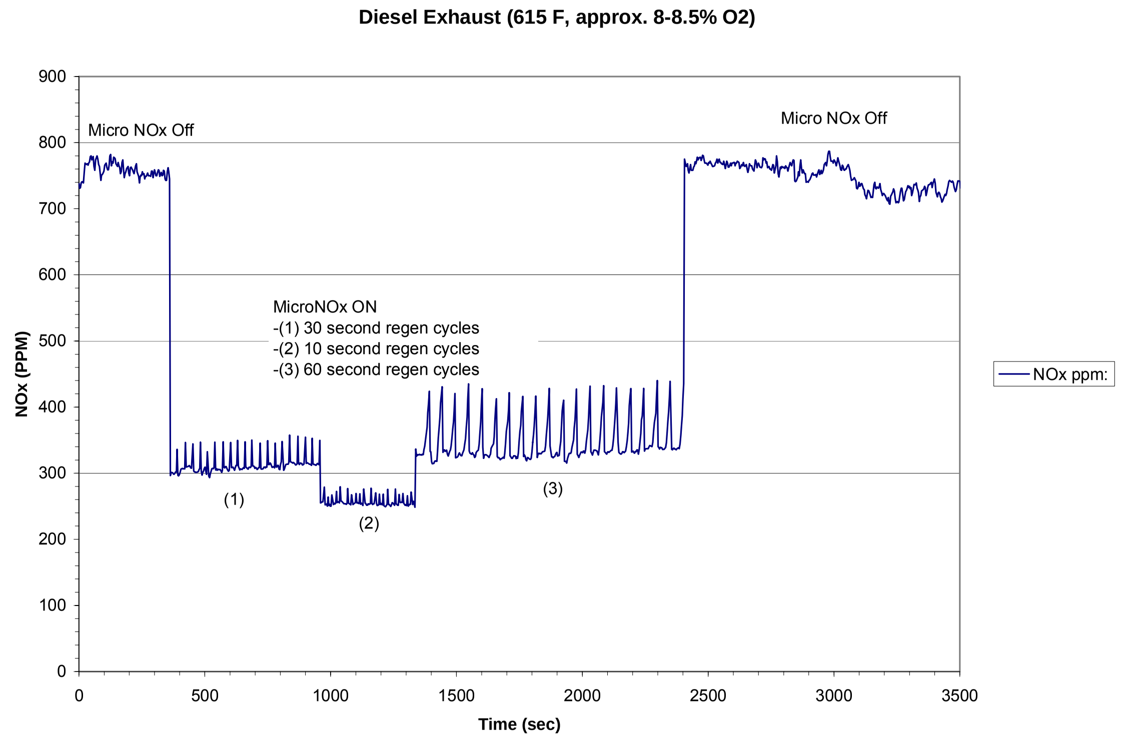


Figure 25. Test Data From MicroNOx system From Cummins B6 Engine.

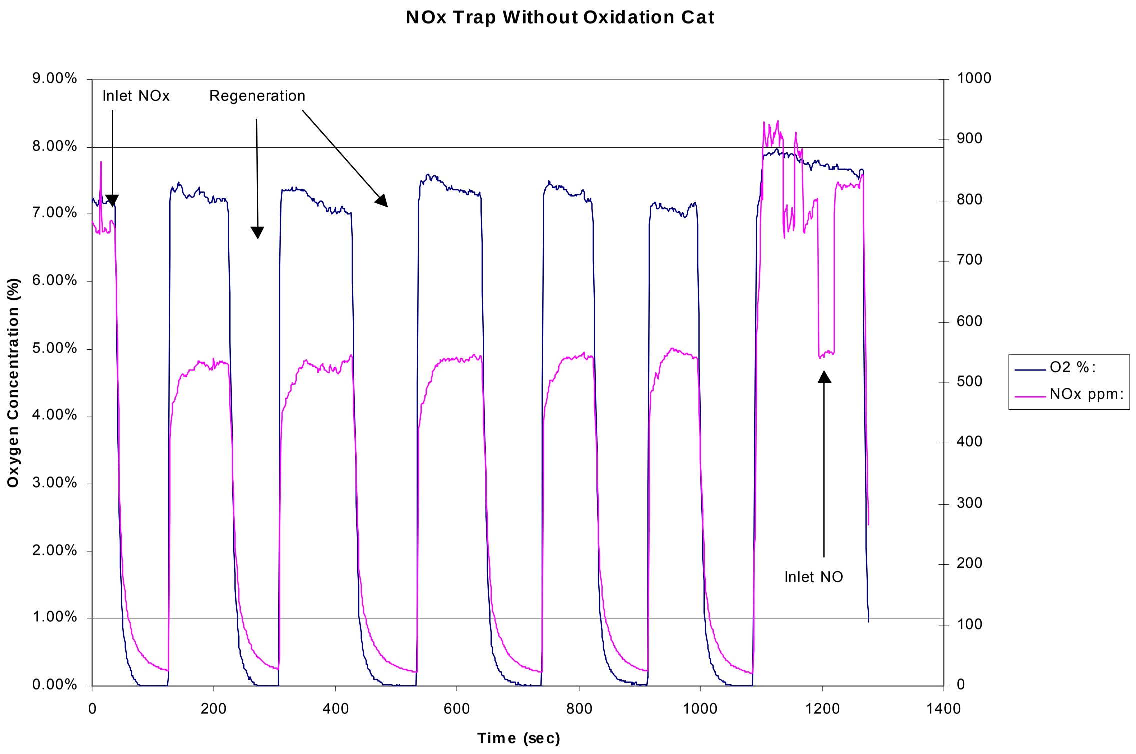


Figure 26. Regeneration and Cyclic NOx Storage Of Catalyst.

Tests of system performance were conducted at MEI with a commercial 20 kW Onan natural gas generator shown in Figure 27. The system was used to evaluate catalyst performance with oxygen levels up to 7% with and without hydrogen generation. The generator was operated at stoichiometric conditions and extra oxygen was injected into the exhaust to simulate a lean burn natural gas engine. Using this generator a single converter consisting of an Pt oxidation catalyst, BaC catalyst, and 3-way catalyst sections. During the regeneration period the flow was by passed. Figure 28 shows performance of the system with 10 second generation cycles. The system produces very high NOx reduction with up to 4% oxygen in the exhaust and then the conversion decreases to the 60 to 70% range at 7%. These results were consistent with the diesel engine results at approximately 8% oxygen in the exhaust. In comparison, the testing on the conventional three-way catalyst showed almost no NOx conversion beyond 1% oxygen in the exhaust.

Figure 27. 20 kW Natural Gas Generator With Test Converter.

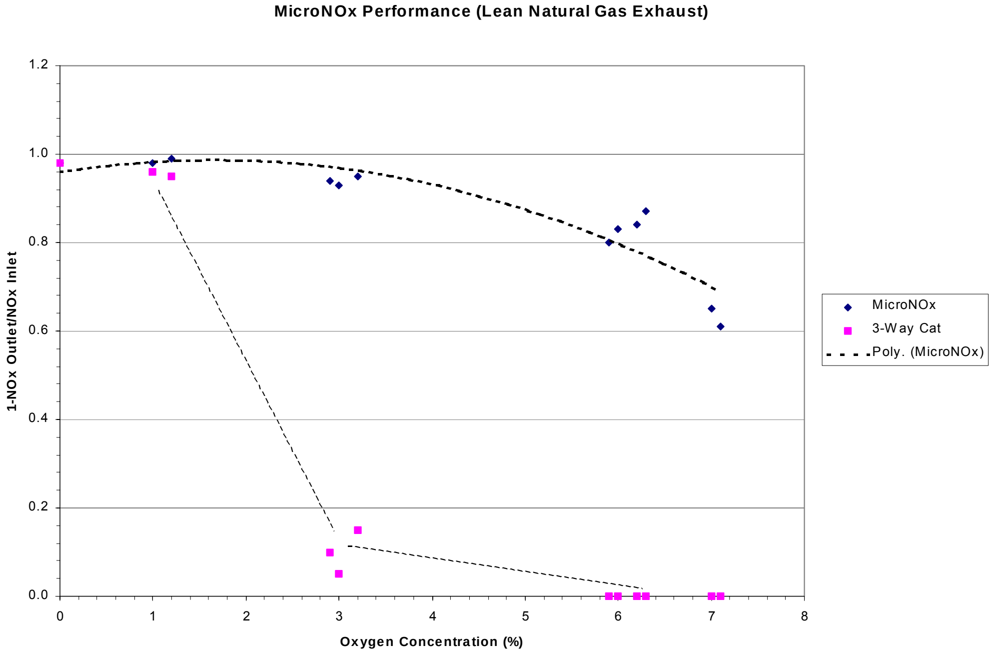


Figure 28. Comparison of MicroNOx System and Conventional Three Way Catalytic Performance In Lean Exhaust Conditions.

4.0 Field Testing

The project culminated in Task 3.0 Field Testing. Task 3.0 consisted of testing using a 100 kW lean burn natural gas set. MEI obtained the generator and coordinated installation at Butte Community College in Chico to operate the waste treatment facility. During the field test the following was accomplished:

- System integration into the field demonstration facility including natural gas and electrical hookups.
- Installation of requisite test and measurement equipment for real time in situ performance measurements.
- Testing and data collection of all system parameters including characterization of NOx levels under various system loads and other operating conditions
- Management of all issues to characterization and implementation of required engineering for real world system operation. (i.e. generator synchronization, mechanical interfaces, instrumentation, continuous operation)
- System design iteration to reflect evolution of catalyst from generation I to generation II catalyst. Based on real world system performance.

4.1 Field Test Setup

A 100 kW natural gas generator set was acquired installed for field testing. Figure 29 shows the generator set and the MicroNOx exhaust system for the unit.

Figure 29. 100 kW Natural Gas Generator Set and MicroNOx Exhaust System For Field Testing

The field test site for the 100 kW generator was a water pumping system at Butte Community College. In discussions with several potential industrial test sites lead to the conclusion that the water pumping station at Butte College would best serve the needs of the program. Butte-Glenn Community College located between Chico and Oroville provides its own wastewater treatment due to its rural location. This system is located on their 900-acre campus, and relies on an aerobic digester system and a series of evaporation ponds. A site map is in shown in Figure 30. The raw sewage is pumped into a pumping well and run through two 5 hp sewage grinders. Two 7.5 hp lift pumps (Figure 31) feed the sewage into an aerobic digester (Figure 32). Two 25 hp blower (Figure 33) aerate the sewage in the digester system. These blowers and pumps run continuously alternating between electric motors. The processed sewage is released into a gravity flow system, pouring into a series of holding ponds where it evaporates. A backup 175 kW generator is stored in the pump house. The test generator replaces an existing back power diesel generator and operated in an “island mode” through a switching system.

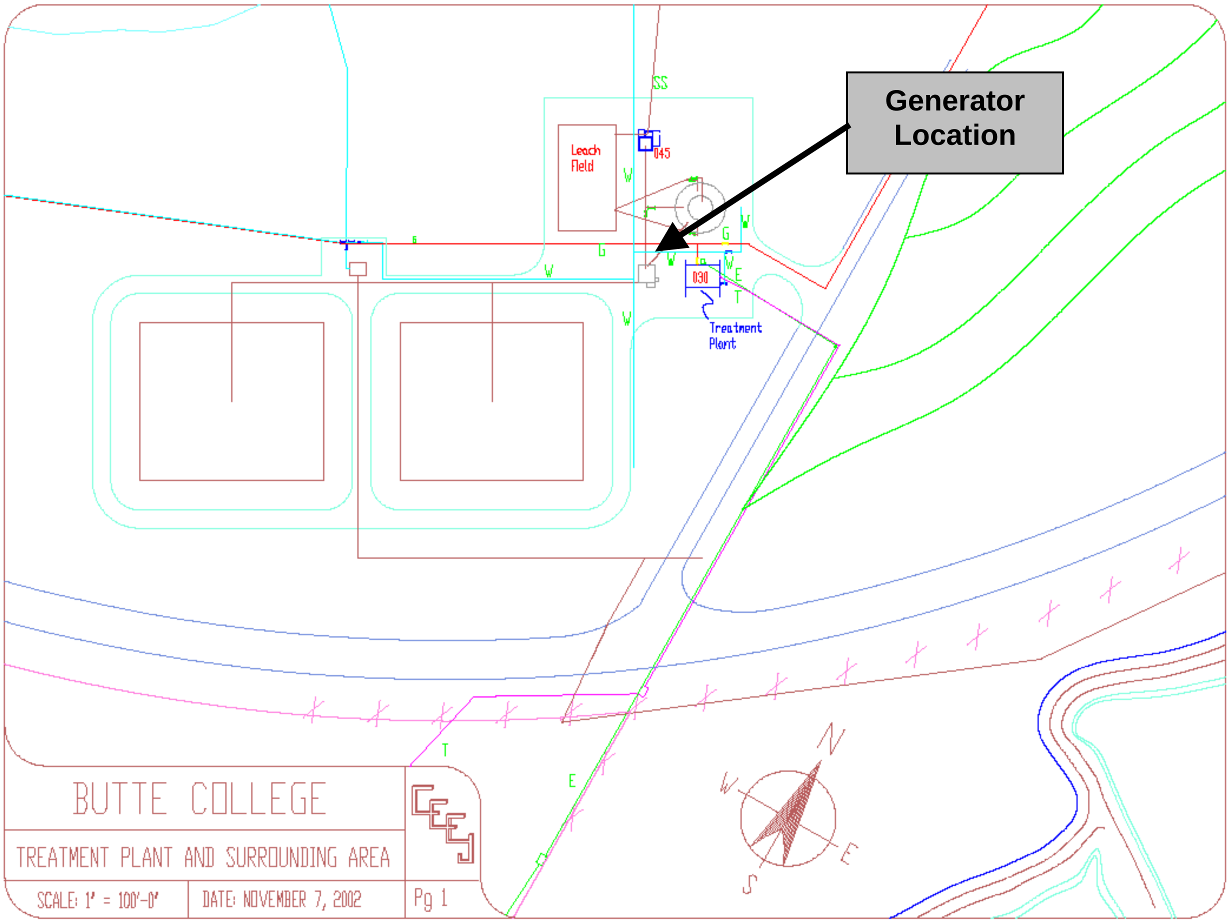


Figure 30. Treatment plant and surrounding area

Figure 31. Lifter Pumps.

Figure 32. Aerobic Digester.

Figure 33. 25 Hp blowers (yellow in background) and existing diesel gen set (foreground).

4.1.1 Field Test Demonstration Genset

The generator set consists of a Chevrolet V8 natural gas engine, a New Age Stanford Generator and digital controls from Incorp. The system runs on 5 psig natural gas at approximately 16:1 air to fuel ratio to produce up to 100 kW of power. Figure 34 to Figure 37show the main components of the generator set. Figure 36 shows the engine detail. The engine is a conventional automotive design. It is a 454 in^3 (7.4 L) Chevrolet, 4 cycle, push-rod type. Bore and stroke are 4.25 x 4.00 in. Horsepower is approximately 230 hp. Carburetion is by an EMCO Natural Gas regulator and manifold set constructed for the Chevrolet V-8. As a stationary power plant it has been modified with non-standard (small valve) heads to run efficiently at a constant 2200 RPM. Cooling is by an oversized radiator. Intake temperature is controlled by a small refrigeration unit that limits cabinet temperature. Table 5 lists engine specifications. Figure 37 shows the Stanford generator and Table 6 lists the generator specifications.

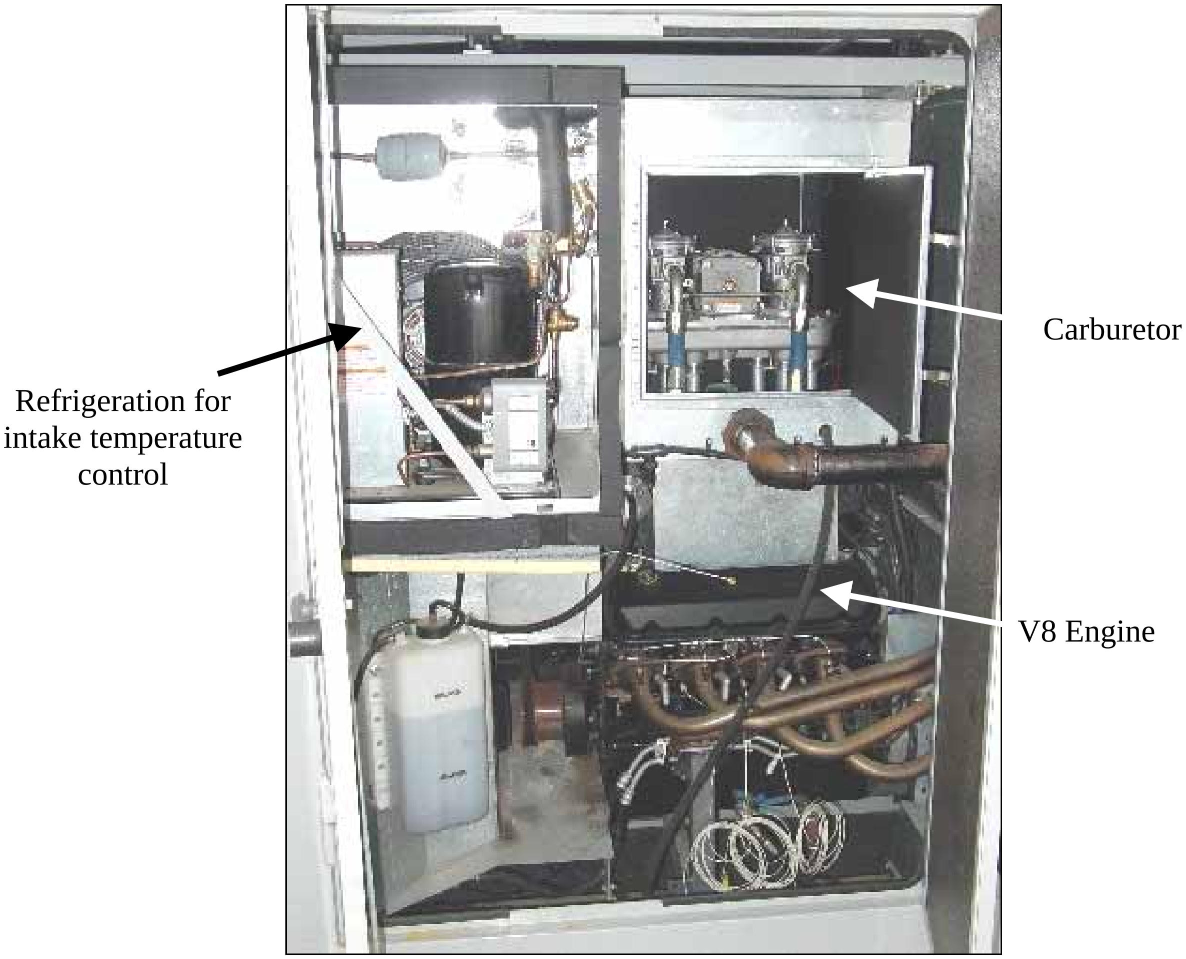


Figure 34. Engine, Carburetor and Refrigeration

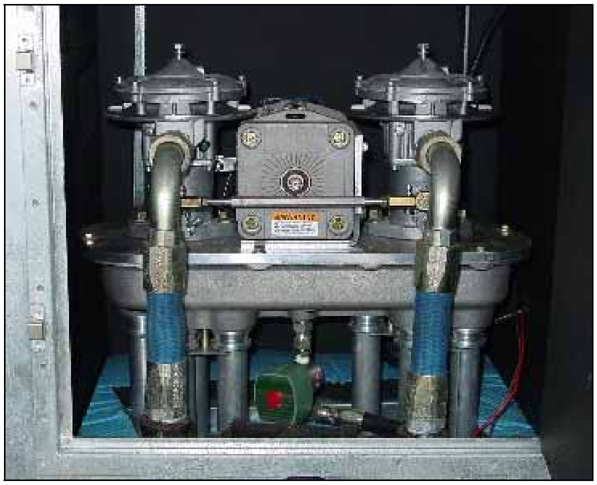


Figure 35. Carburetor Close Up

Figure 36. V8 Engine Close Up

Table 5. Natural Gas Engine Specifications

Manufacturer	Chevrolet
Fuel	Natural gas (5 psig)
Configuration	V8
Displacement	7.44 L (454 in^3)
Valves	2 per cylinder (16 total)
Bore	108 cm (4.25 in)
Stroke	102 cm (4.00 in)

Figure 37. New Age Stanford Generator

Table 6. Generator Specifications

Manufacturer/Model	New Age Stanford
Power Output	100 kW
Speed	1800 RPM
Current	150 Amp continuous

The 100kW generator set install at Butte Community College is shown in Figure 38. The NOx reduction system is mounted on the top of the generator as shown in Figure 39. The control system for actuating valves, monitoring exhaust temperature, and operating the reformer is mounted inside the generator enclosure as shown in Figure 41. The NOx content in the exhaust is measured by a Rosemount Analytical NOx analyzer that is located in the build adjacent to the generator as shown in Figure 42. The NOx analyzer can be set to sample from either upstream or down stream of the catalyst system by means of a three-way valve. Calibration gas for zero and span checks on the analyzer are located in the building with the analyzer.

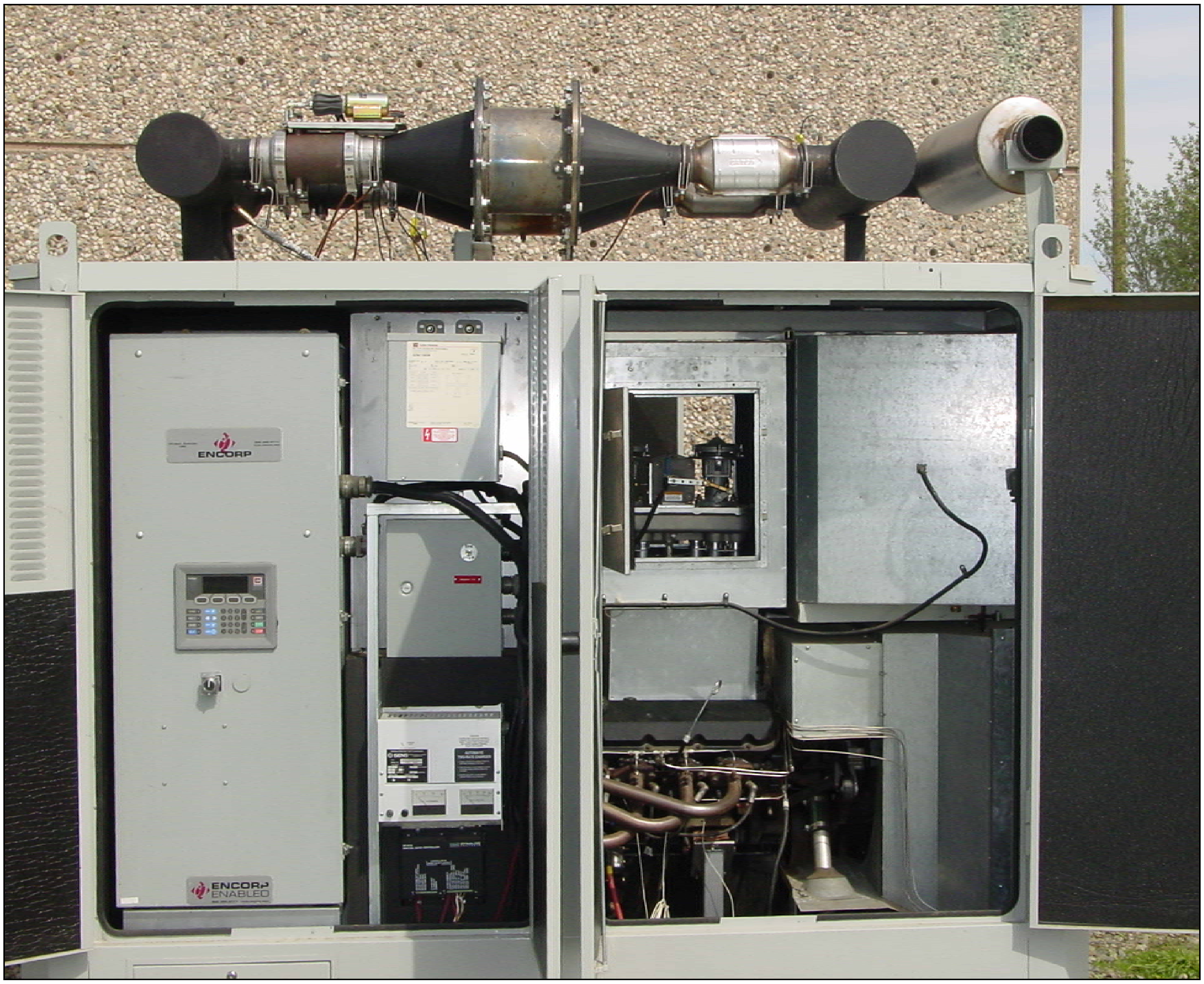


Figure 38. MicroNOx Exhaust System Installed For Field Testing.

Catalysts for the NO oxidation and NOx storage were prepared as described in section 2.3.7. The catalyst was prepared in quarter sections and confined with a retaining ring. A typical catalyst section is shown in Figure 45.

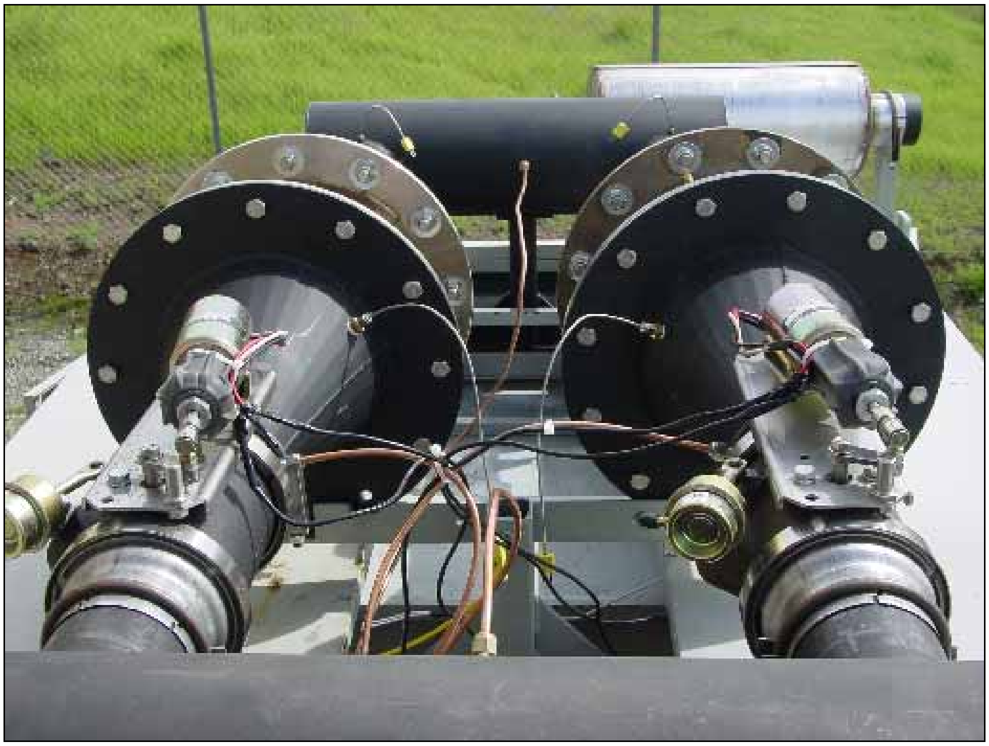


Figure 39. NOx Reduction Catalytic Sections And Exhaust Flow Control Valves.

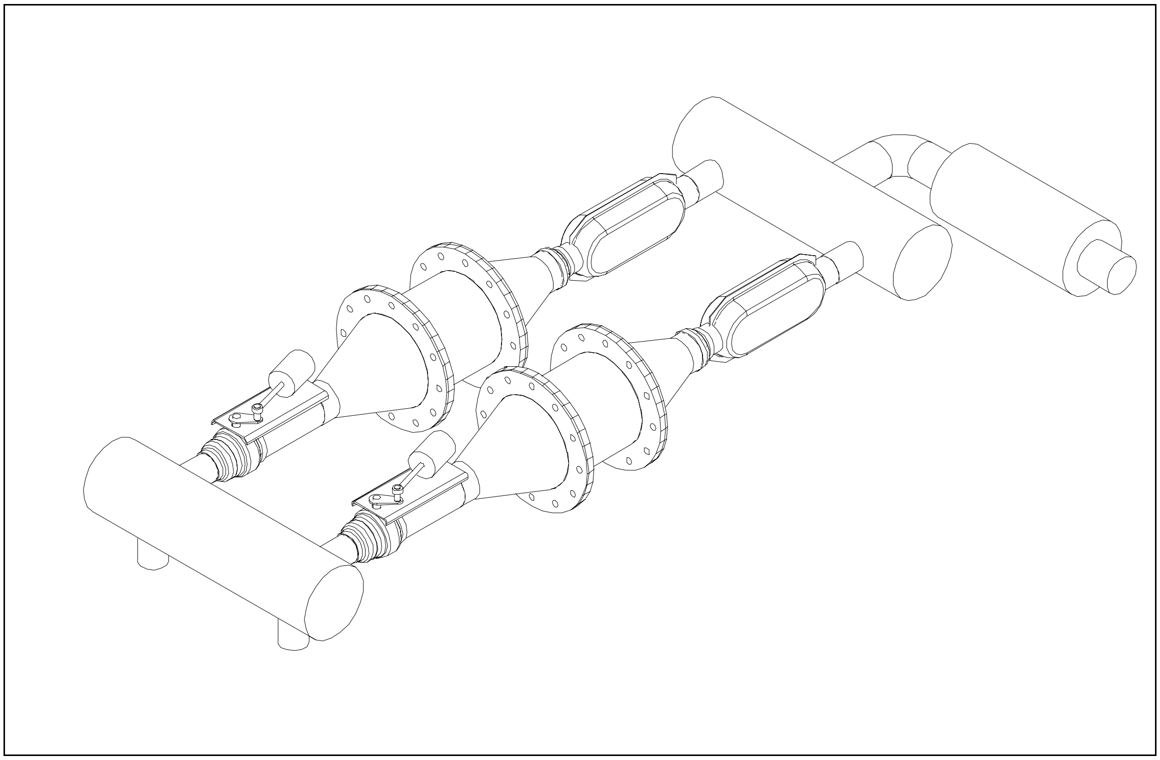


Figure 40. 3D CAD Drawing of the MicroNOx Exhaust System

Figure 41. Exhaust Temperature Instrumentation and controls.

Figure 42. NO/NOx Analyzer Instrumentation.

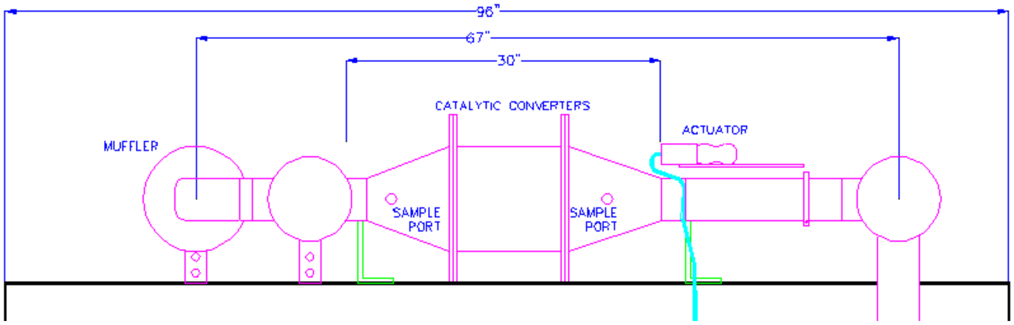


Figure 43. MicroNOx Exhaust System Line Drawing

Figure 44. MicroNOx Exhaust System Field Installation.

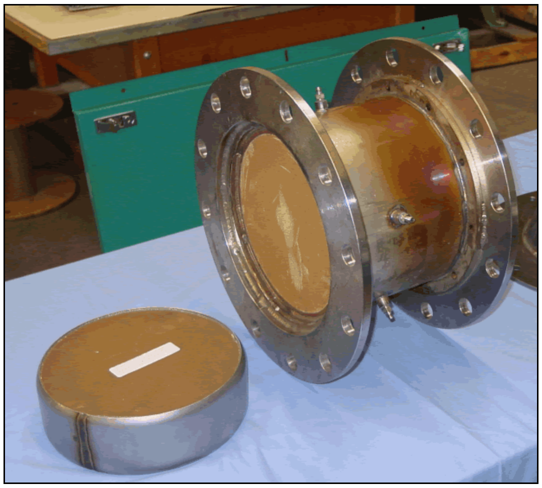


Figure 45. Catalyst Honeycomb Substrate and Catalyst Installed in System

4.1.2 Methane Reformer

Figure 46 shows the methane reformer. The reformer converts methane and air into hydrogen and carbon monoxide. To provide stable flow, the reformer gas supplies to the reformer use sonic orifices. With sufficient gas inlet pressure, the flow is independent on the pressure variations downstream of the orifice.

While the reformer is equipped with a heater and closed loop temperature control, the reformation of methane is an exothermic reaction, once it is initiated. A possible modification of the system is to take advantage of the exhaust heat to initiate the reaction, eliminating the need of a heater.

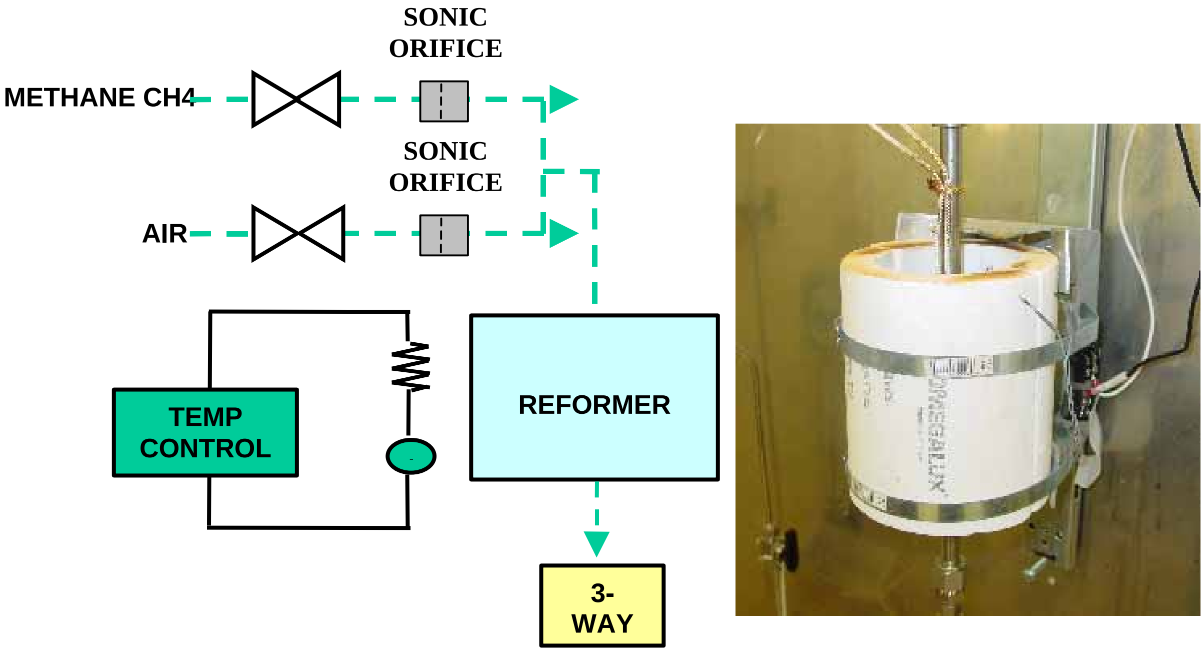


Figure 46. Methane Reformer

4.1.3 Electric Switch Gear

Makel Engineering’s generator operates offline through a switching system located in the electronics building. It enhances operations by providing power for the digester system. This saves Butte College the cost of the equivalent power purchased from PG&E. Figure 47 illustrates the switching system located at the pumping station. Figure 48 shows the digester pumps and Figure 49 shows the generator with switching hardware.

Figure 47. Treatment Plant Switching System

Figure 48. Treatment Plant Digester System Pumps

Figure 49. System and Electrical Swithcing Hardware

4.1.4 Field Demonstration Test Results

Initial check out testing of the system resulted in over 400 hours of operation with conditions ranging from 0% to 12% excess oxygen and generator loads from 20 to 90%. A summary of the mapping data collected during the reporting period is show in Figure 50. The data show high level of NOx reduction (greater than 90% conversion) out to approximately 6 to 7% oxygen in the exhaust. The plot shows different generation times and regenerator flow rate. The NOx conversion is more sensitive to regeneration time than to the flow rate over the range tested. Regeneration times greater 60 seconds decreased conversion of NOx.

During the survey testing, each leg of the device was monitored periodically. This testing revealed that the right side of the system showed less NOx reduction than the left side. While the cause is not yet known, it may be flow leakage around the catalyst or reduction in effectiveness of the catalyst.

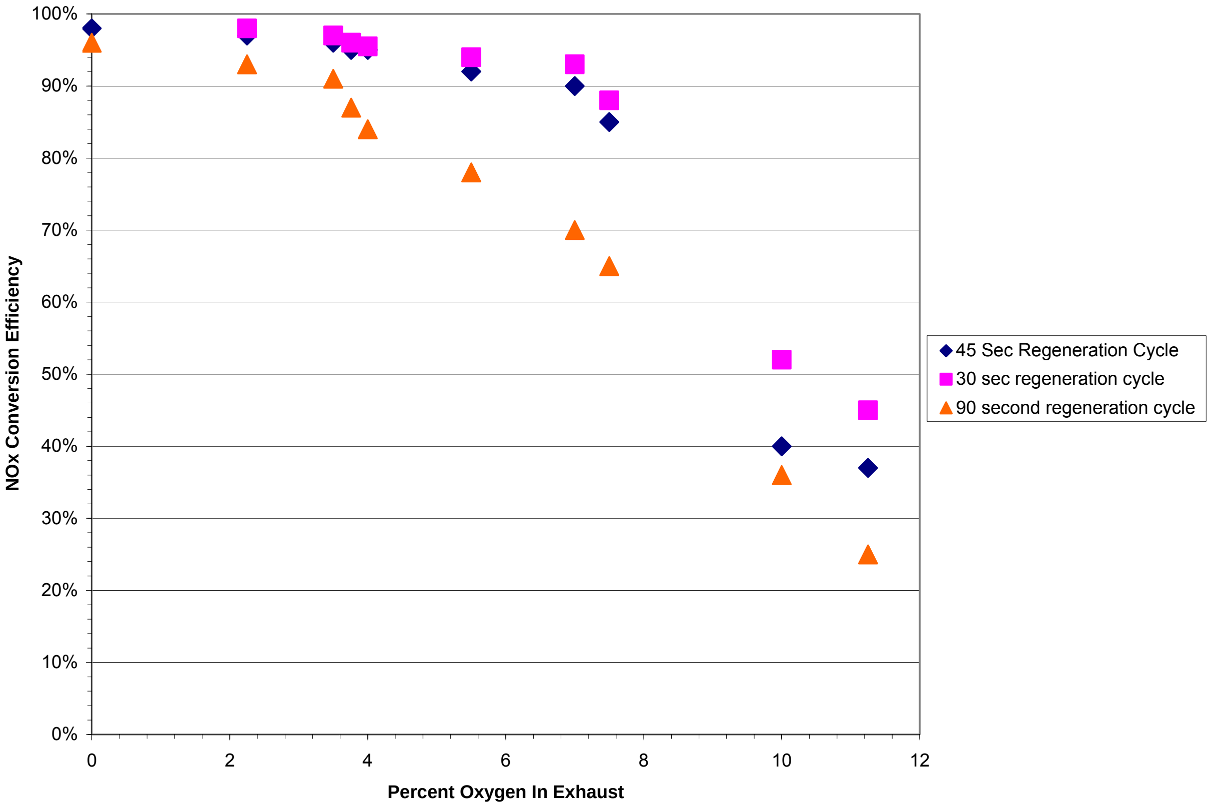


Figure 50. NOx Conversion As A Function Of Percentage Oxygen In Exhaust and Regenertion Cycle Time

Figure 51 shows the NOx levels upstream (out of the engine) and downstream (after treatment) of the catalytic converter with approximately 4% excess oxygen in the exhaust stream. The incoming NOx concentration is approximately 200 to 250 PPM. On the downstream side of the converter the NOx level is measured to be less than 10 PPM. The decay of the NOx signal switching from upstream to downstream is due to limitation of the pumping capacity of the NOx analyzer. Regeneration gas flow rate was 200 cc/min into the converters during testing. Under these conditions the system exhibits very high NOx reduction. A preliminary mapping of NOx reduction versus exhaust oxygen concentration has been performed. Figure 52 shows operation of the unit with approximately 11% excess oxygen in the exhaust. The conversion efficiency of the system is seen to drop to approximately 40%. A summary of the mapping data collected during the reporting period is show in Figure 50. The data show high level of NOx reduction (greater than 90% conversion) out to approximately 6 to 7% oxygen in the exhaust.

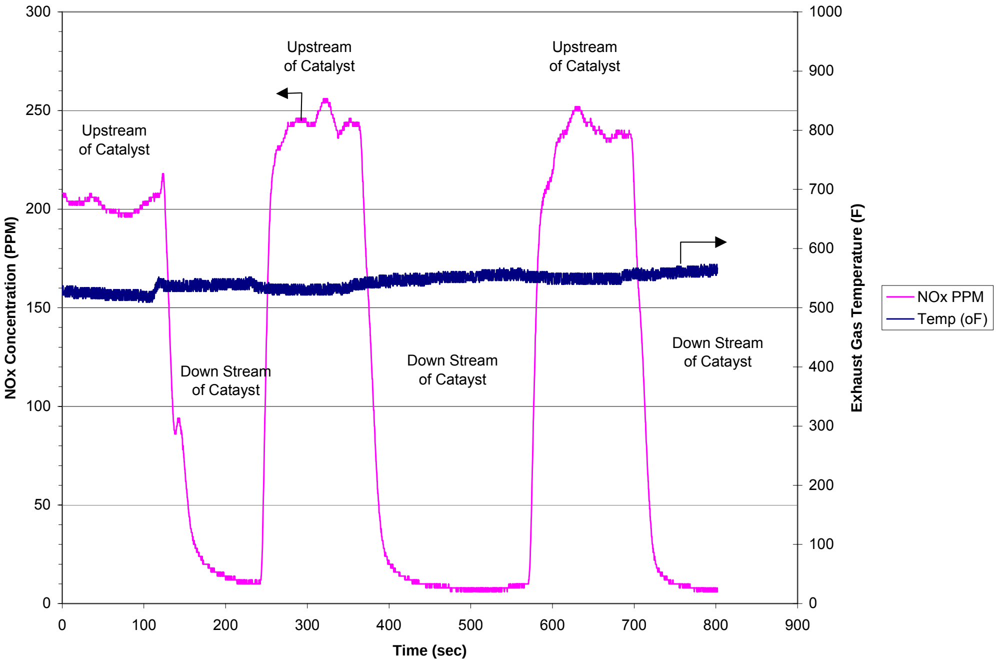


Figure 51. Data Showing Comparison of NOx Upstream Of NOx Reduction System and Downstream of NOx Reduction System With 4% Excess Oxygen in Exhaust.

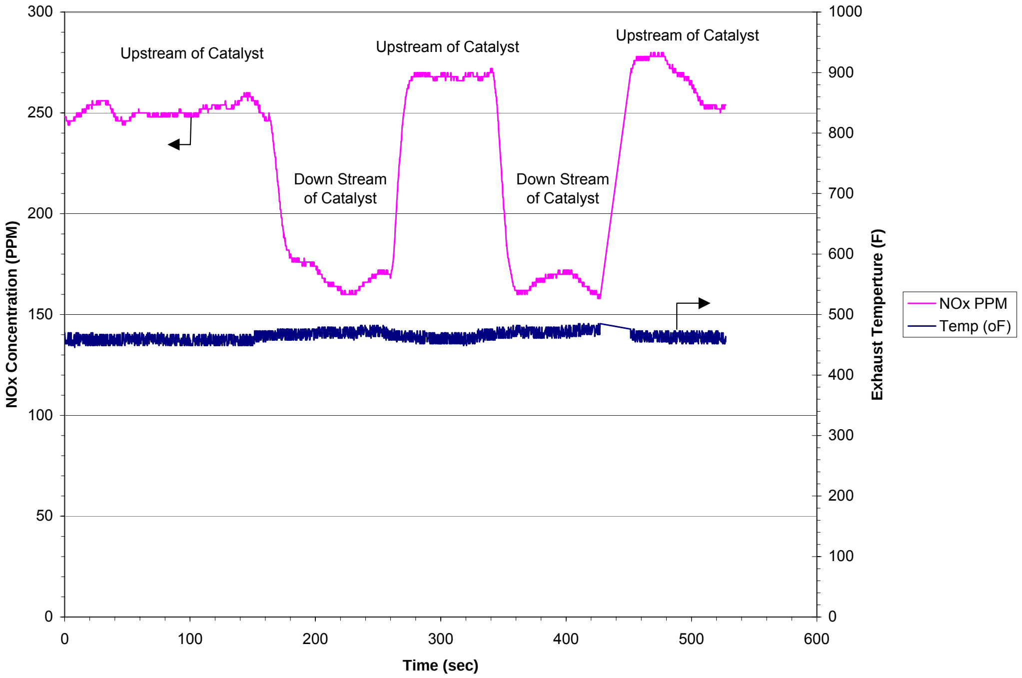


Figure 52. Data Showing Comparison of NOx Upstream Of NOx Reduction System and Downstream of NOx Reduction System With 11% Excess Oxygen in Exhaust.

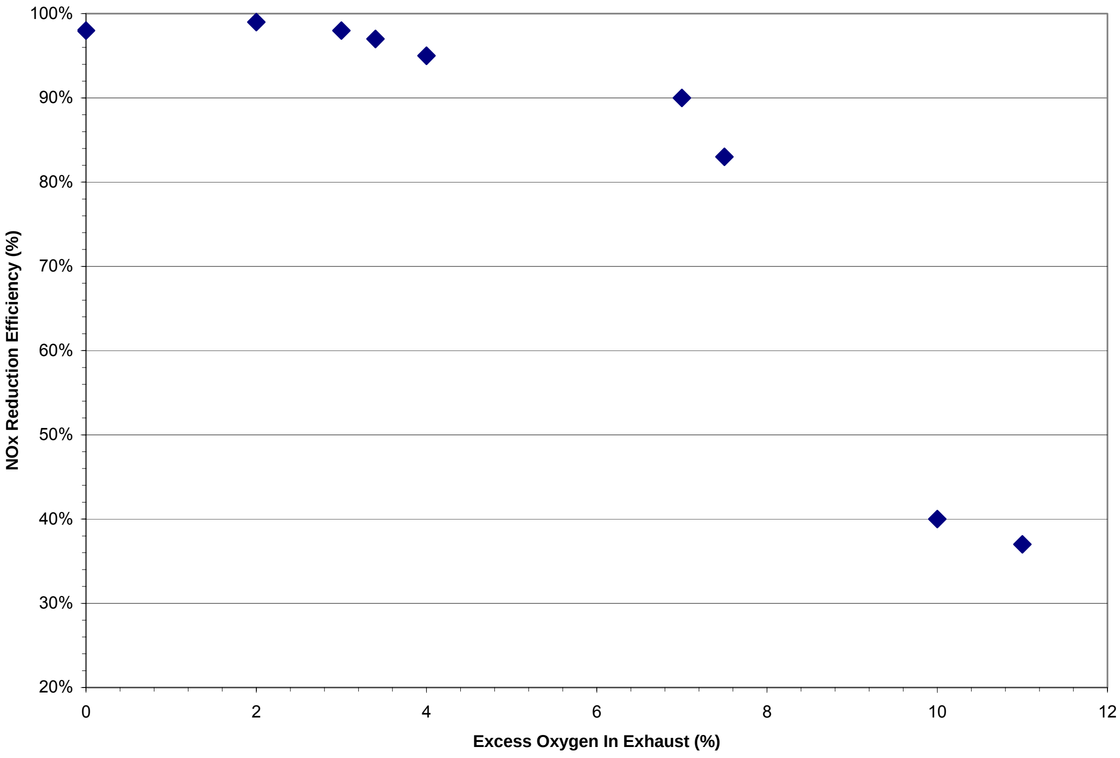


Figure 53. NOx Conversion As A Function Of Percentage Oxygen In Exhaust.

Best operating mode for the field testing is listed in Table 7. At this condition the majority of field testing was conducted. Engine air/fuel ratio was controlled by throttling the inlet fuel to the engine with external valving. A condition of 6% oxygen in the exhaust stream was chosen as it provided the most stable operating condition for operation of the facility. While survey tests were conducted at up to 12% oxygen in the exhaust stream, the gen set was unable to produce sufficient power for the peak load of the water treatment facility. The system was set for operation over a six month period of time to be switched on to operate the facility. Measurement of the NOx concentration upstream and down stream of the system were obtained and used to calculate NOx conversion efficiency.

Conversion efficiency = 1- NOx(outlet)/NOx(inlet)

During the course of testing the output of the reformer was periodically sampled to verify the production of CO and H2 for the regeneration. Sample bottles were prepared and evacuated at MEI and taken the Butte College test site. The product from the reformed was collected in bottles with condensate removed and the samples were analyzed with a Tracor 540 gas chromatograph. During the course of the testing no degradation of the reformer performance was observed.

Table 7. Best Operating Mode.

Gas inlet temperature	325 to 360 C
Oxygen concentration in exhaust	4%
RPM	1800
Reformer fuel flow	0.045% of total
Reformer equivalence ratio	2.5 to 3.0
Regeneration time	60 seconds

Extended testing of the MicroNOx system at the BCC test site indicated a degradation of system performance over time. After 200 hours of operation a drop in NOx reduction from approximately 84% to less than 75% was apparent with a trend downward. The possible reasons for this drop in performance were identified as (1) physical degradation of the catalyst section leading to exhaust flow by pass, (2) contamination of the catalyst with sulfur or other fuel based contaminate, or (3) unknown deactivation of the one or more of the catalyst sections.

The system was disassembled and inspected which revealed small channels were segments of the catalyst were pieced together Figure 54. These channels would not occur if the large catalyst substrates were prepared a single piece rather than the quarter sections that were processed due to limitations of the chemical processing equipment. Additionally samples of the natural gas at the test site were sampled and analyzed for sulfur content. The results from the sample indicated a total sulfur content less than 20 PPM due to H_2S and odorant, therefore deactivation due to sulfur is not suspected as the degradation problem.

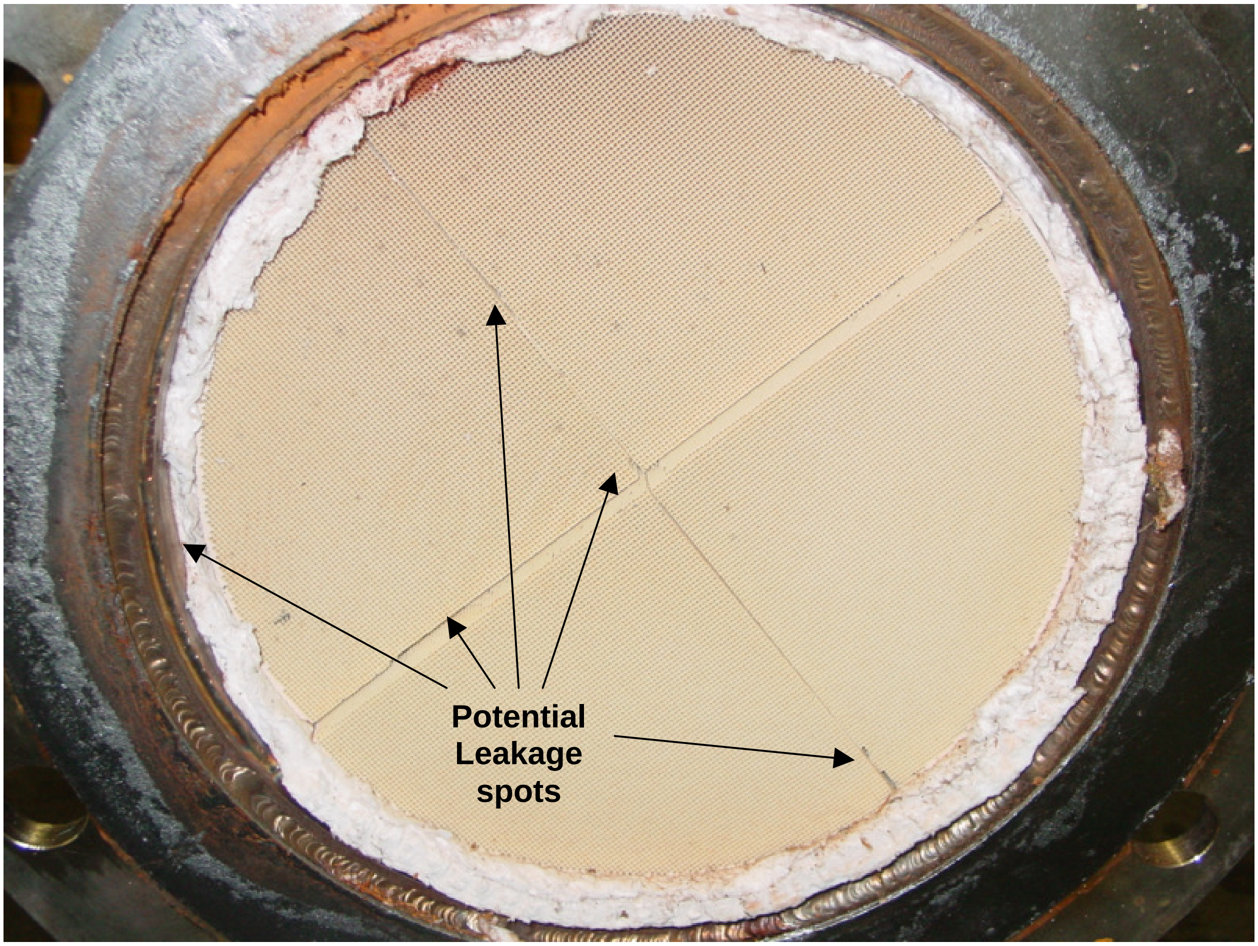


Figure 54. Post Test inspection of NOx Trap Catalyst section indicated potential leakage sites.

To address the problem a new catalyst preparation protocol was developed with increase the barium carbonate and platinum loading. The catalysts were prepared and sealed into a welded catalytic converter housing shown Figure 55.

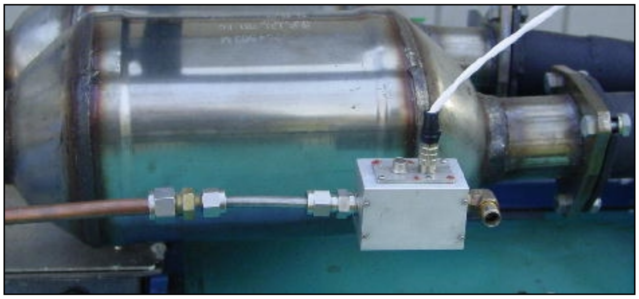


Figure 55. Modified Catalytic Converter with Improved Catalyst and Production Packaging.

The field test system was upgraded with two of the modified converters and testing resumed. The system with modified converters are shown in Figure 56, Figure 57, Figure 58, and Figure 59.

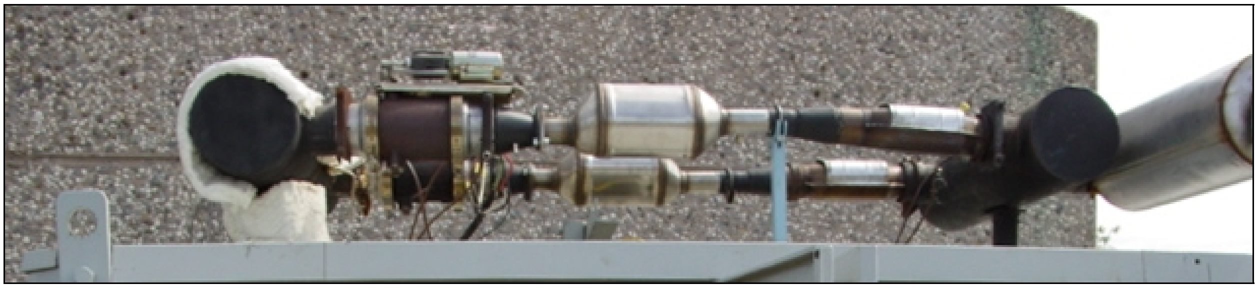


Figure 56. MicroNOx Exhaust System with Secondary Treatment System.

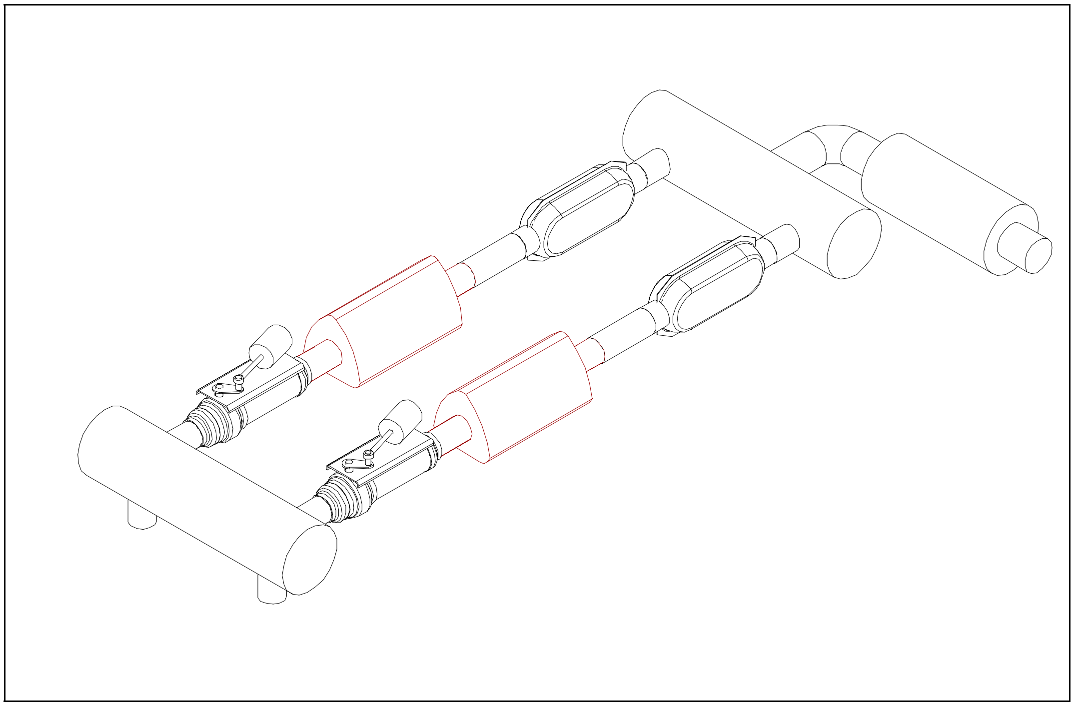


Figure 57. MicroNOx Exhaust System with Secondary Treatment System.

Figure 58. MicroNOx Exhaust System 3 way catalytic converter Treatment System.

Figure 59. MicroNOx Exhaust System Installed For Field Testing.

The degradation of the initial catalyst and the steady performance of the subsequent catalyst is shown in Figure 60.

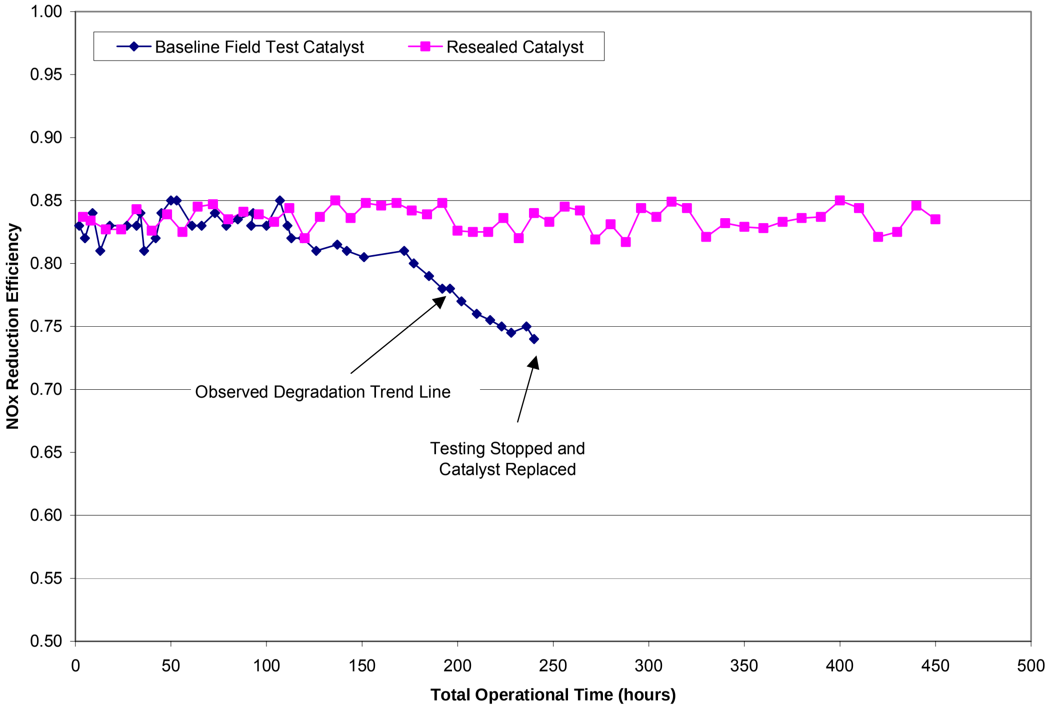


Figure 60. MicroNOx System Performance With Initial Catalyst And With Resealed Catalyst.

5.0 Conclusions and Recommendations

Subsequent to the initiation of this project changing market, conditions have precipitated the need to revisit some assumptions related to the commercial potential of this technology. While the energy crisis of a few years ago appeared to mark the beginning of a trend towards the rapid growth of distributed power generation, in fact, the market has not expanded as rapidly as was expected and the current market climate is one of generally stable energy prices and availability. Where once a rapid growth of distributed power generation catalyzed by the ongoing risks of both unstable prices and uncertain reliability was expected, customers cost and reliability expectations have been reasonably met and they have not been driven by economic necessity to implement distributed power solutions. An additional market dynamic that has changed over the past few years is the emergence of catalytic and low temperature combustion micro turbines. These systems are capable of operating with very low NOx levels and therefore do not require additional exhaust after-treatment. These substantial changes have refocused market development efforts on a class of applications that represent a significant commercial opportunity and represent a substantial benefit to both the market and greater community.

Market research results have identified Combined Heat & Power (CHP) as the new class of application poised to benefit from the technology developed through this research. CHP, often called cogeneration, because it combines the generation of electricity and heat energy from a single fuel source is a compelling value proposition to virtually every hotel, semiconductor FAB, bottling plant, food processing facility, hospital, wood products mill, etc. who uses large amounts of utility-supplied electricity and natural gas for use in steam boilers or water heaters. In cogeneration systems, natural gas is diverted into an engine to generate electricity. Then, the waste exhaust heat is used to make steam or hot water in a Heat Recovery boiler, which can then be used for heating or cooling (via heat absorption chilling) purposes. This simple system is typically 40%-60% more efficient than traditional energy generation and delivery methods. Combined Heat & Power systems are not new, and have been proven in thousands of applications worldwide. By adding a NOx reduction after-treatment system that mitigates high levels of pollution, these systems can be implemented more readily in compliance with local air quality standards throughout the United States.

Of the CHP/cogeneration market there is a special subset of bio-gas fueled applications that are particularly suited to adopt this technology including animal waste, agricultural organics, and forest/urban green waste. Each of these bio-gas fueled applications represent a substantial untapped customer base able to adopt a new cost saving and environmentally friendly technology. Facilities in each of these application areas have a need for both utility-supplied electricity and natural gas for use in various operations.

- In the case of landfills, energy is used for plant operation and wastewater evaporation
- In the case of dairies, energy is used to both operate and clean the facilities.
- In the case of agricultural facilities, energy is used to process and store the food products.
- In the case of forest waste, energy is used to process and dry the wood products.

The most mature of these application areas is Landfill systems. In California today, 70 landfills are currently collecting bio-gas and simply flaring the gas with no additional benefits. These 70 landfills have the potential for producing approximately 66 MWe of electricity. We have identified the three largest landfills currently flaring more than 5 Mwe. The next eight medium sized landfills (1-5 MWe) have approximately 14 MWe of potential electrical capacity, and the remaining 59 flaring landfills have approximately 21 Mwe of generation capacity. Providing a clean burning low cost gas conversion technology that meets ARB clean air standards will unlock this potential resource saving these landfills a substantial energy expense and leverage a resource that is currently wasted.

5.1 Commercialization

Commercialization of the technology will be pursued through joint product development with stationary power genset manufacturers. It appears possible to produce a system that can sell for less than $15/kW for systems over 100 kW. Ultimately, this technology may be applicable to mobile sources with the focus on large engines. Because of the cost and the difficulties in introducing new technology into the transportation market, near term market objective is stationary sources.

5.2 Recommendations

This program demonstrated a novel NOx aftertreatment system that is suitable for use on lean burn natural gas or propane engines. Overall NOx reduction exceeding 80% was demonstrated with the system with a fuel consumption penalty of less than 0.5%. The technical work performed on this project suggests several improvements to the design used for this pilot demonstration and extensions of the technology to expand its market potential.

Specific recommendations to advance the technology to commercialization are:

- Improve catalyst processing to enable large substrates to the processed as single pieces.
- Investigate methods of integrating the reformer directly with the catalytic converter to take advantage of waste heat from converter and reduce parts.
- Extend the technology to diesel fuels addressing sulfur content, particulate tolerance, and elevated inlet temperature.
- Continue development and improvement of NOx storage catalyst, improving storage characteristics at elevated oxygen concentration (i.e. leaner conditions).

6.0 References

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Ref. 9 Influence of sulphur content in fuel on the fuel consumption and pollutant emissions of vehicles with gasoline and diesel engines (1999)

Appendix A Design Package