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USE OF A DIESEL FUEL PROCESSOR FOR RAPID AND EFFICIENT REGENERATION OF SINGLE LEG NOX ADSORBER SYSTEMS

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ABSTRACT

Lean NOx adsorber systems are one of the primary candidate technologies for the control of NOx from diesel engines to meet the 2007-2010 US emissions regulations, which require a 90% reduction of NOx from the 2004 regulations. Several of the technical challenges facing this technology are regeneration at low exhaust temperatures and the efficient use of diesel fuel to minimize fuel penalty. A diesel fuel processor system has been developed and tested in a single leg NOx adsorber configuration on a diesel engine test stand. During NOx adsorber regeneration, this fuel processor system performs reduces the exhaust O_2 level to zero and efficiently processes the diesel fuel to H_2 and CO. Combined with a NOx adsorber catalyst, this system has demonstrated NOx reduction above 90%, regeneration of the NOx adsorber with H_2/CO pulses as short as 1 second and fuel penalties in the 3 to 4% range at 50% load. This fuel processor system can also be used to provide the desulfation cycle required with sulfur containing fuels as well as providing thermal management for PM filter regeneration.

INTRODUCTION

Emissions regulations in the US for 2007-2010 require a reduction in the NOx emissions levels from 2.0 g/bhp-hr to a level of 0.2 g/bhp-hr. While in-cylinder advances have been able to reduce NOx emissions by significant levels, from pre-regulation levels above 15 g/bhp-hr to current 2002/04 levels of 2 g/bhp-hr, a further reduction of 90% is generally considered to require after-treatment controls. In addition, it is generally considered necessary for such after-treatment controls to reduce NOx by at least 80% and preferably 90%. NOx adsorber or NOx trap technology is considered one of the after-treatment technologies that could meet these stringent levels of NOx control. Numerous reports have documented the ability of NOx adsorber systems to reduce NOx emissions by over 80%. For example, Blakeman, et.al. show engine test data in which a NOx adsorber system decreases exhaust NOx levels by 95% above an exhaust gas temperature of 300°C with the NOx conversion decreasing rapidly below 300°C (Blakeman, 2003). In other tests, NOx conversions >80% could be demonstrated from 210°C to about 400°C. Below the temperature ranges listed, the NOx conversion dropped quickly. Mital, et.al. showed similar results with >90% NOx control above about 270°C low fuel injection rates and above ~220°C with much higher fuel injection rates (Mital, 2003). A dual leg system with two parallel NOx adsorber systems was tested by Schenk and Laroo on a heavy-duty diesel engine (Schenk, 2003). This two leg systems allows the one leg to be operated with greatly reduced exhaust flow during regeneration allowing reduced fuel consumption. Good performance was demonstrated at higher exhaust temperatures, > 80% NOx conversion above 290°C. Numerous other reports show clearly that exhaust NOx can be reduced by >90% in engine tests.

For NOx adsorber systems to achieve the required level of NOx control and overall engine performance, several operating conditions must be achieved.

- The NOx adsorption capacity must be sufficiently high to adsorb all of the NOx produced by the engine during the lean adsorption portion of the cycle. While the NOx adsorption capacity can be increased by increasing the size of the NOx adsorber, this can lead to packaging, weight and cost issues for the vehicle system. In addition, maintaining a high NOx adsorption capacity requires full regeneration of the NOx adsorber during the rich cycle.
- Reduction of the NOx to N_2 and regeneration of the NOx adsorber requires that the environment around the NOx adsorber become reducing, that is that the O_2 level is zero and a reducing agent be present. Reduction of exhaust oxygen can be done a number of ways. The engine can be throttled, EGR levels can be increased and fuel can be injected to raise the fuel air ratio to the stoichiometric point. While engine operating strategies that produce stoichiometric or rich exhaust mixtures are feasible, some of these strategies produce high smoke levels,

cause high soot formation within the cylinder and can be detrimental to engine durability.

- Good NOx adsorber operation over the entire exhaust temperature range is required to achieve the high level of NOx control required. Diesel fuel is a good reductant for the NOx adsorber system at temperatures above 300°C but below this temperature it is not sufficiently reactive to regenerate the NOx adsorber. More reactive reductants can be generated by partially combusting the diesel fuel within the cylinder using "late cycle injection" strategies where the high temperatures in the cylinder partially oxidize the fuel to make H_2, CO and other reactive small hydrocarbons.
- Fuel sulfur can reduce NOx adsorber capacity and must be periodically purged or separately trapped. Desulfation strategies require raising the NOx adsorber temperature to above 500°C and in some reports to above 700°C and providing rich conditions to purge the sulfur from the NOx adsorber catalyst. Achieving such high temperatures is generally done by combusting diesel fuel on the NOx adsorber catalyst and then driving the exhaust mixture rich to desulfate the adsorber catalyst.

Several literature reports show that more reactive reductants can regenerate NOx adsorber systems at low exhaust temperatures (Mital, 2003; Schmolz, 1993), typically reporting NOx adsorber regeneration below 200°C in engine tests and as low as 150°C in laboratory tests. The wide operating temperature range and the potential for other benefits led

DESIGN CRITERIA FOR THE DIESEL FUEL PROCESSOR

The Diesel Fuel Processor (DFP) described in this report is developed to address many of the issues related to achieving the desired NOx adsorber performance. The specific performance goals for the DFP unit are:

- Use diesel fuel as only required input
- Volume of 0.5 to 1 liter per liter of engine displacement
- Convert diesel fuel with high efficiency to very reactive reductants, H_2 and CO
- Require that the engine only reduce exhaust O_2 level to ~5 to 8% during the regeneration cycle to limit demands on the engine
- Allow efficient NOx adsorber regeneration over the entire exhaust temperature range of interest, 150°C to > 500°C
- Provide thermal management by producing high exhaust temperatures, >700°C if needed, for desulfation cycles or PM filter regeneration
- Aid in NOx adsorber desulfation by producing the required conditions to purge sulfur

ENGINE TESTING STRATEGY AND FACILITIES

Engine testing was conducted on several turbo charged and after cooled compression ignition engines, a 7.2 liter Caterpillar and an 8.3 liter Cummins. These engines had NOx emissions levels in the range of 3 to 4 g/kW-hr. All testing was done on a diesel fuel processor unit with an effective volume of 6.3 liters. This resulted in DFP to engine swept volume ratio of 0.88 to 0.76. Within this range, performance of the diesel fuel processor was relatively similar for these engines. These engines were fitted with intake air throttles to allow control of the exhaust oxygen during the regeneration cycle. The general system configuration is shown in Figure 1. The DFP was located between the turbo charger outlet and the inlet to the NOx adsorber catalyst. The DFP fuel injection was controlled by a separate microprocessor based control unit. This DFP control unit was interfaced with the engine control unit (ECU) to allow coordination of the engine operation with DFP operation. The NOx adsorbers were third party units and were sized to the engine emissions by the suppliers.

Exhaust gas samples for emissions measurements were take upstream of the DFP for engine out emissions, just downstream of the DFP to monitor the effectiveness of fuel processing and downstream of the lean NOx adsorber to measure engine out emission. Gas sampling lines and intermediate filters were heated above 200°C to keep water and hydrocarbons (including uncombusted diesel fuel) in the gas phase.

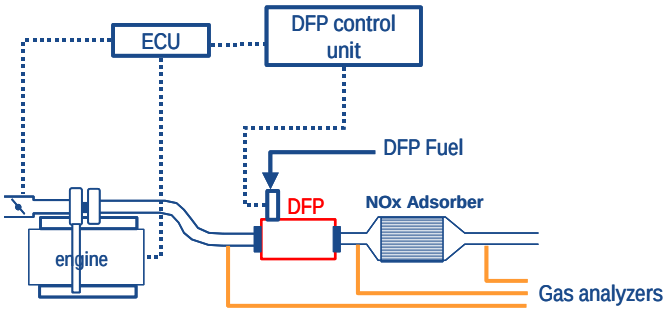

Figure 1. Configuration of after-treatment system for tests.

The analyzer system consisted of typical gas analysis instruments located in a separate room with a gas sample transit time in the range of 15 to 30 seconds. Where needed, the analyzer time base was shifted by a set time period to correct for different transit times to each instrument. An example where this was necessary was for the analytical results for H_2 and CO. In one particular test where H_2 was

measured, a special purpose mass spectrometer was used to measure H_2 concentration downstream of the DFP. This instrument was installed close to the engine on a separate sampling line and the response was delayed only 1 to 2 seconds compared to the remaining gas components with a transit delay of about 30 seconds.

DEMONSTRATION OF DIESEL FUEL PROCESSOR PERFORMANCE

To demonstrate operation of the DFP, the exhaust was sampled downstream of the DFP unit as shown in Figure 1 and a regeneration cycle initiated. Test were done at steady state engine conditions. Prior to each test, engine throttle conditions were established to produce an exhaust O_2 level of ~5% with added fuel to maintain engine torque and rpm. These throttle and engine fuel conditions were then used for the NOx trap regeneration cycle at this load condition. Exhaust emissions results just downstream of the DFP unit are shown in Figure 2 during a typical regeneration cycle at 50% load conditions. The trace at the top of the figure shows the appropriate throttle schedule for this data set. The fuel schedule to the DFP was programmed to produce a rich pulse of 3 seconds duration. The data of Figure 3 show a H_2 and CO production with a concentration peak of 3.5% H_2 and 3% CO with pulse widths of about 3 seconds at half height. These concentrations are measured at the analyzer and with pulse broadening in the sampling line, the maximum concentration in the exhaust stream could be higher. The 1% minimum oxygen levels is due to mixing in the sample line or slow analyzer response; other tests have shown that the O_2 during the rich pulse is zero.

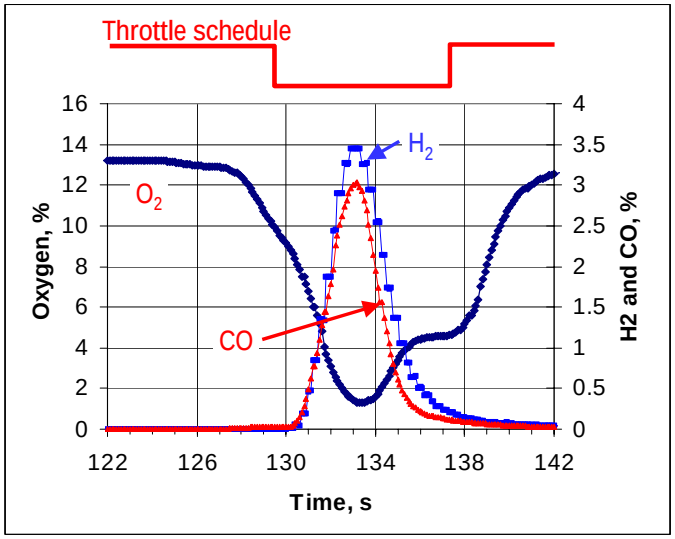

Figure 2. Demonstration of DFP operation. Exhaust H_2, CO and O_2 levels measured downstream of the DFP unit during a typical regeneration cycle.

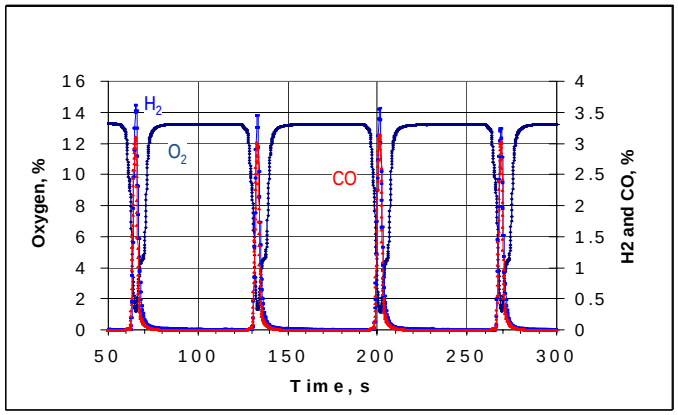

Figure 3. Exhaust gas composition downstream of the DFP unit during repetitive 60 s lean trapping and 3s rich regeneration cycles.

The engine/DFP system was programmed with a repetitive cycle of 60 seconds lean operation followed by 3-second rich regeneration of the NOx adsorber. The exhaust composition upstream of the NOx adsorber is shown in Figure 3.

During regeneration, the additional fuel required in the cycles described above can be divided in three categories:

- Additional fuel required by the engine due to throttling to reduce the exhaust O_2 level—As the engine is throttled, the engine will require additional fuel to maintain torque. This fuel must be included in the total required for NOx adsorber regeneration. The fuel efficiency effectiveness between post cycle injector in-cylinder injection or throttling to reduce exhaust O_2 level is not well established and should be explored.
- Fuel required to consume the remaining oxygen in the exhaust—The exhaust O_2 level must be reduced to zero and the fuel to do this can be either injected during in-cylinder combustion, injected post cycle using engine fuel injectors or in-pipe using downstream injectors. To reduce the O_2 level in the exhaust, essentially all these methods are equivalent since the stoichiometry of the reaction is defined.
- The fuel required to produce H_2/CO for the regeneration of the NOx adsorber—Again, whether done in-cylinder or within the exhaust pipe or in a downstream device such as the DFP, all such systems are equivalent. The only difference is in the efficiency or in the ability of such systems to effectively and efficiently

produce H_2 and CO. The system with the most effective conversion of fuel to H_2 and CO will be preferred.

Fuel economy of the system is directly related to the ratio of lean time to rich time required by the NOx adsorber catalyst. The tradeoffs between these parameters can be assessed independently and will be discussed below. The cycle described in Figure 2 required a given amount of fuel for the regeneration cycle that produces that level of H_2 and CO compared to the level of fuel consumed by the engine during normal lean operation. If the additional fuel required by the rich NOx adsorber regeneration is determined and compared to the fuel required during normal engine operation, then the fuel penalty can be determined. The fuel penalty is defined as the additional fuel required for 1) engine throttling, 2) fuel to react with the remaining O_2 in the exhaust during the rich cycle, and 3) fuel to produce the H_2 and CO. Using a theoretical model for fuel consumption and the test data from the on-engine testing, a semi-empirical model was developed for fuel consumption during the NOx adsorber cycle. The fuel penalty calculated in this manner is shown in Figure 4.

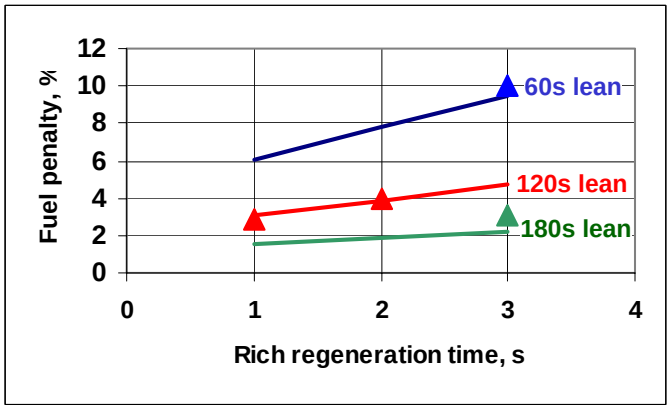

Figure 4. Fuel penalty associated with various lean NOx adsorption times and rich reforming times for the DFP system.

The data from the engine test at 60 second lean time and 3 second rich regeneration time is shown as the solid triangle in the data of Figure 4 on the 60s lean curve. Tests at other engine conditions are also shown in Figure 4 as additional solid tiangle data points. As the reforming pulse time is decreased, the fuel required decreases as does the fuel penalty. Similarly, as the lean time is increased thus decreasing the frequency of the rich regeneration cycles, the fuel penalty decreases as well. With lean times of 120 seconds and with reforming time of 2 seconds, the fuel penalty can be in the range of 4% for the DFP-NOx trap system described here. Shorter lean times raises the fuel penalty and longer lean times decreases the fuel penalty. Similarly, shorter reforming times reduces fuel penalty and longer reforming times increase fuel penalty. Several additional engine test data points are included on Figure 4 as X points at various lean times and reforming times.

The required lean time and rich regeneration time for good NOx emissions reduction is determined by the performance of the NOx adsorber catalyst. If the NOx adsorber catalyst can effectively utilize the H_2 and CO generated by the DFP, then the amount of H_2 and CO required can be small. If the utilization of reductant is poor, then the amount of reductant required could be large. The effective reductant production is shown in Figure 5 where the ratio of reductant produced by the DFP to the NOx in the exhaust is graphed versus lean time and rich regeneration time similar to Figure 4. These data are an extrapolation of actual integrated H_2 and CO measured in the engine tests.

Theoretically, with equal amounts of NO and NO_2 in the exhaust, and with stoichiometric reaction between the reductants and the NOx, the required ratio of reductant to NOx would be 1.5. This required level is represented by the dashed line in Figure 5. As can be seen, all of the points for 60 and 120 seconds lean time and greater then 1 second rich regeneration time have a ratio of reductant to NOx higher then this minimum required ratio. Thus, if the NOx adsorber can efficiently utilize the H_2 and CO and efficiently adsorb the NOx, then this system can achieve very good fuel economy.

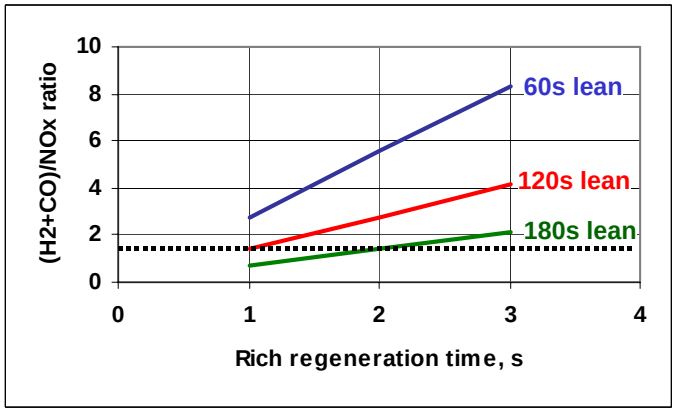

Figure 5. Ratio of reductant (H_2+CO) produced to NOx in the exhaust for various lean and rich cycle times.

The DFP and NOx adsorber system was operated at steady state conditions and 50% load for several lean times and rich regeneration times over a number of cycles until the NOx level in the exhaust had reached a steady state value. The overall NOx conversion and fuel penalty was then measured. These data are summarized in Table 1. Fuel

penalties in the range of 4% were measured with a corresponding NOx conversion in the range of 90%.

Table 1. Fuel penalty and NOx conversion measured in engine tests for various lean and rich times.

Lean time	Rich time	Fuel penalty	NOx conversion
s	s	%	%
60	2	7.3	>95
120	3	4.5	~92
120	2	3.8	~87

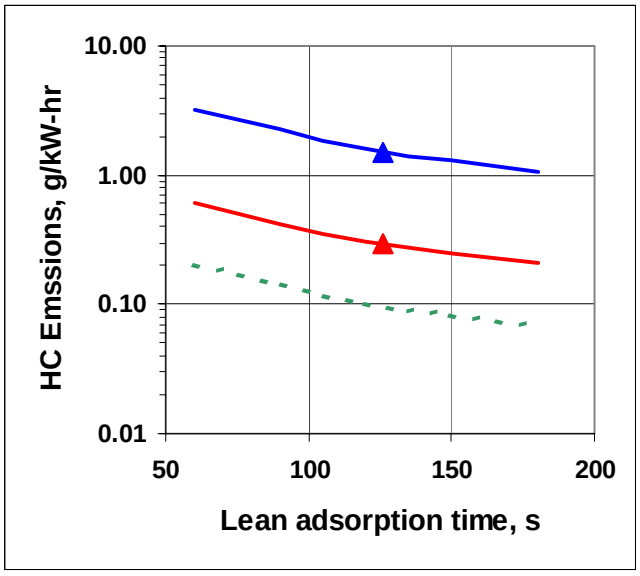

Figure 6. Hydrocarbon breakthrough from the DFP during operation.

Again at 50% load, hydrocarbon emissions downstream of the DFP were measured. Actual measured values are shown as triangles in Figure 6 and the effect of lean adsorption time on this HC emissions level is shown as the extrapolated theoretical line. The different curves are for different catalyst designs. The first generation system showed an HC emissions level in the range of 1.8 g/kW-hr while a second-generation catalyst reduced this level to 0.3 g/kW-hr. An advanced catalyst system, currently under development, should reduce this level to the range of 0.1 g/kW-hr at 120 seconds lean adsorption times. It should also be noted that in these advanced systems with reduced hydrocarbon breakthrough, a larger proportion of the hydrocarbon emissions is methane. During the engine tests, measurement downstream of the NOx adsorber catalyst showed very low unburned hydrocarbon levels.

Diesel fuel contains sulfur and this sulfur will react on the NOx adsorber catalyst decreasing the NOx adsorption capacity. This sulfur must be periodically purged from the NOx adsorber catalyst to maintain the NOx capacity. Since the sulfur level in the fuel is low, in 2007 and beyond it will be 15ppm, the effective sulfur poisoning is small, resulting in a need to desulfate the NOx trap every 300 to 500 miles. The issue is that desulfation of the NOx adsorber catalyst requires a high temperature, typically 600 to 750°C. The high temperature desulfation has been identified as the major cause of thermal deactivation of the NOx adsorber and strategies for regeneration that reduce the thermal stress on the adsorber could increase system durability.

The DFP system has been designed to be placed upstream of the NOx adsorber catalyst and to be used to raise the exhaust gas temperature and heat the NOx adsorber catalyst to the desulfation temperature. When the NOx adsorber is at the desired desulfation temperature, the DFP will produce active reducing agents. The data for a subscale DFP performing such a desulfation cycle is shown in Figure 7. The DFP operates in thermal management mode to raise the exhaust temperature to 600 to 650°C. After some period, when the NOx adsorber is sufficiently hot, the DFP can be used to generate H_2/CO pulses of any desired duration for effective regeneration.

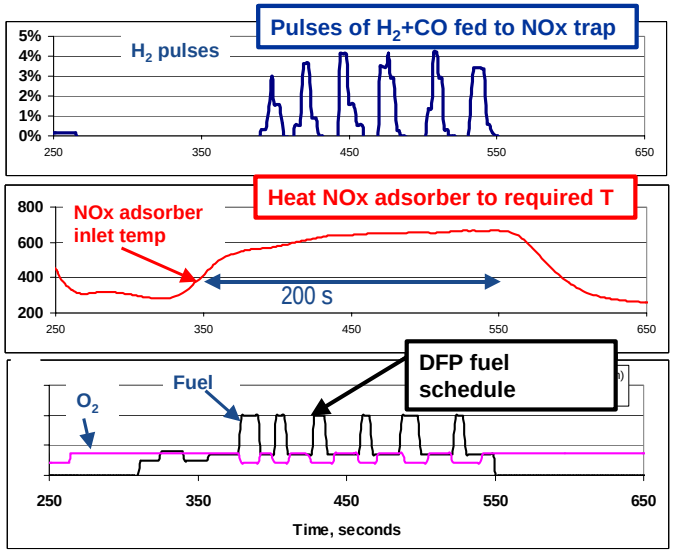

Figure 7. Subscale rig demonstration of a potential NOx adsorber desulfation cycle.

SUMMARY

A diesel fuel processor unit has been developed to enable optimum operation of single leg NOx adsorber catalyst systems for the control of NOx emissions from diesel engines. This unit has been applied to heavy-duty diesel engines configured with NOx adsorber catalysts to demonstrate operation. Work to date has demonstrated operation over a broad range of exhaust temperatures, from 180 to 360°C, and has shown >90% NOx control with low fuel economy penalty. This diesel fuel processor also provides thermal management capability, that is, can be used to heat the exhaust gas and down stream components to temperatures above 700°C. This capability is useful in NOx adsorber desulfation strategies. Such a desulfation strategy has been demonstrated on subscale.

Work is ongoing to further develop this system and to optimize the performance of the combined diesel fuel processor and NOx adsorber catalyst to obtain high levels of NOx control, low fuel penalty and low overall system cost, size and weight. The optimum system will most likely require a NOx adsorber catalyst that is designed specifically for this diesel fuel processor and for the very reactive reductants produce by the diesel fuel processor.

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