DIESEL ENGINE NOX REDUCTION VIA NITROGEN-ENRICHED AIR*

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Abstract

It has long been known that NOx emissions from diesel engines can be reduced by reducing the peak temperatures of combustion. Dilution of the cylinder charge with inert gases is one method of lowering the peak cylinder temperatures. Nitrogen is an obvious diluent; however until recently its use was limited to stationary laboratory engines. Compact, high productivity, air separation membranes have recently been developed. These membrane modules provide means for generating nitrogen-enriched air (NEA) at the point of use, for example, under the hood of a diesel truck. NEA offers an attractive, clean alternative to dilution with exhaust gases.

NEA is generated by feeding the cooled, turbocharged air to the bore side of a hollow fiber membrane device. A pressure differential across the wall of the hollow fiber causes oxygen to permeate preferentially through the polymeric wall. Thus as air flows along the length of the hollow fibers, it becomes slightly depleted of oxygen and enriched in nitrogen. The resulting NEA is fed to the intake manifold of the engine at only slightly lower pressure than the turbocharged air. The oxygen-enriched co-product (OEA) is simply vented to the atmosphere. The effectiveness of the NEA for NOx emissions reduction is also related to the composition of the NEA. Only slight enrichment is needed and NEA compositions in the 80% to 82 % nitrogen range prove to be very effective in NOx reduction.

Developments of the NEA technology have progressed beyond the laboratory engine scale. NEA is now being studied on a number of commercial engine platforms with good success. NOx emission reductions as high as 50% are being achieved on diesel engines supplied with membrane generated NEA. Results from some of these tests are presented.

Introduction

Air separation membranes have become a practical means for on-site generation of nitrogen during the last 20 years. Major industrial gas companies utilize highly selective polymeric membranes for the removal of oxygen from compressed air to produce and deliver on-site

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pressurized, high purity nitrogen gas. In the screening of polymers for such membranes it was recognized that there is a trade-off between selectivity (the relative rates at which gas species permeate across the membrane) and the permeability or the gas flux across the membrane.

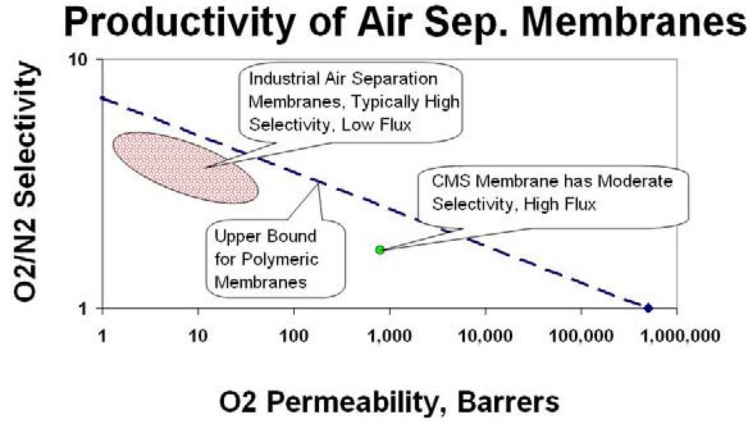


Figure 1 Relationship between selectivity and permeability in polymeric air separation membranes.

Robeson observed that there appears to be an upper limit to polymeric membrane selectivity, the ability to distinguish between two gas species, that declines with increasing permeability, the rate at which a gas will transport through the membrane barrier. Figure 1 shows an empirical fit to the highest oxygen-nitrogen selectivity observed for polymers with increasing permeability for oxygen. As such, Robeson's Line seems to represent the natural or fundamental limits of air separation in polymeric membranes. When producing high purity nitrogen, it is desired that the membranes have the highest selectivity in order to maximize the amount of nitrogen produced from the air feed. Such membranes fall within the upper left region in Figure 1. To compensate for their low membrane flux, high purity nitrogen generation units are typically operated at high pressures (100 to 200 psig). Even so, the surface area of such membrane required would be such that they could not be fitted under the hood of a truck! By contrast, CMS membrane polymers lie in the mid-range and though less selective are more productive, having oxygen fluxes that are over two orders of magnitude higher than those in commercial industrial air separation membranes. Owing to their high flux, it is also possible to operate such membranes at lower pressure, thereby reducing the amount of compression energy necessary to operate the membrane. As will be shown later, the diesel engine application discussed here requires only modest nitrogen enrichment in the intake air to have a dramatic effect on NOx emissions. Thus the combination of low compression energy, high membrane flux, and low NEA concentrations are all favorable for the diesel engine application of membranes of polymers in the latter region of Figure 1. The perfluoropolymer membrane developed by Compact Membrane Systems (CMS) and their partner Innovative Membrane Systems (IMS) provides the added features of chemical resistance and robustness needed for operation under the hood of a truck.

The diesel engine industry has been facing increasingly stringent regulations and exhaust emissions limits. Table 1 shows the emissions requirements for on-road diesel engines that come into effect in 2007. Substantial reductions in both NOx and particulate matter (PM) are required to meet increasingly tough emissions legislation by the US Environmental Protection Agency and the California Air Resource Board in 2004 and beyond.

Table 1. History of Diesel Truck Engine Exhaust Legislation

Emission Component	**U.S. 1998**	**U.S. 2002/04**	**U.S. 2007**
NOx Emission (g/bhp-h)	4.0	2.0	0.2
PM Emission (g/bhp-h)	0.1	0.1	0.01

Exhaust gas recirculation (EGR), a cost-effective way of reducing NOx levels from spark-ignition engines, is now being implemented on compression-ignition engines [1-3]. However, its applicability continues to be under scrutiny because of increased particulates (PM) and smoke at higher engines loads [4, 5], possible oil contamination [6] and subsequent wear implications [7], and reduced engine durability [8-9]. Table 2 summarizes and distinguishes the features of the EGR and NEA approaches to diesel engine NOx reduction.

Table 2. Comparison of nitrogen-enriched air vs. EGR for NOx in diesel engines.

Nitrogen Enriched Air	**Exhaust Gas Recirculation**
• Clean, nitrogen-rich air is delivered by a membrane free of intake airborne particulates. • No effect on engine life or durability. • Acts as a heat exchanger to further lower intake air temperature. • Homogeneous mixture makes trade-off among NOx, particulates, and brake-specific fuel consumption predictable. • Requires a membrane separator with compensation for flow loss and for pressure loss.	• Unwanted exhaust species circulated to the intake air and an increase in particulates. • Presence of sulfur and/or carbon affects engine life and durability. • Hot EGR will affect engine power. • Cylinder-to-cylinder variation is difficult to optimize, and trade-offs among NOx, PM, and BSFC are difficult to predict. • Requires EGR cooler with controls and a filter to trap particulates and sulfur compounds.

Many investigators [10-12] have used nitrogen-enriched air to reduce NOx emissions from diesel engines. All of these engine tests were conducted by using bottled pure nitrogen, primarily to simulate the effects of EGR. The effect of depleting oxygen in intake air (or nitrogen-enrichment) can, however, be achieved by using an on-board, air separation membrane [13, 14].

NOx emissions correlate with combustion temperatures reached in the diesel engine cylinder. The flame temperature is dependent on inlet pressure and temperature and on the chemical composition of the fuel and oxidant [15, 16, 17]. The influence of intake air oxygen concentration on the adiabatic flame temperature was examined by Olikara and Borman [18]. Figure 2 shows the adiabatic flame temperature decreases with a decrease in intake oxygen levels at any air-fuel equivalence ratio. When the intake oxygen concentration reduces from 21% to 17%, there is a reduction in the stoichiometric flame temperature of about 250ºK. The combustion models suggest that the combination of lower oxygen concentrations and lower flame temperatures result in reductions in the amount of NOx formed in the combustion chamber.

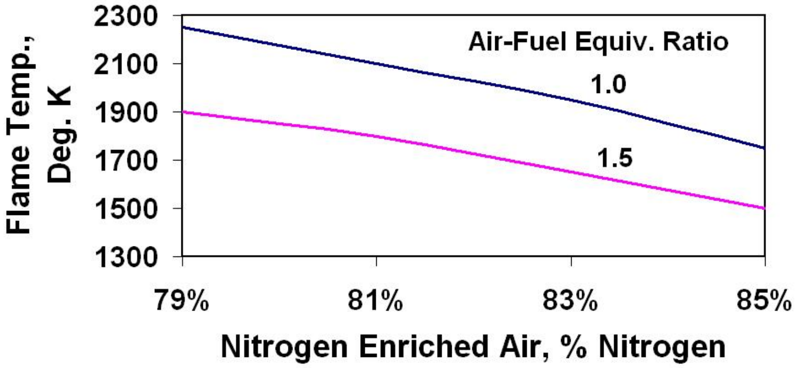


Figure 2. Influence of NEA and Air-Fuel Equivalence Ratio on Adiabatic Flame Temperature

Several investigators, using analytical models and engine tests, have reported that intake air oxygen concentration is the most influential parameter in controlling NOx emissions [10-12, 16, 19]. These studies show the range of excess air factors typical of diesel engine operation can be readily attained with air separation membranes. Onboard membranes can readily increase the nitrogen levels from 79% to 83% in the NEA. Due to the presence of oxygen in the exhaust, comparable adjustment of the oxygen level with EGR requires replacement of a large fraction of the fresh air with EGR to maintain the corresponding oxygen levels. Supplying the desired intake air oxygen concentration with a membrane device could eliminate the use of EGR and many of its limitations listed in Table 2. The supplemental addition of NEA to an EGR engine is also recognized as a means for gaining further reduction in NOx emissions.

<u>MEMBRANE CONSIDERATIONS</u>

The basic principle of membrane operation and various designs and operating characteristics are described in Nemser et al [14]. The hollow fiber (HF) membranes and devices being employed in NEA applications are described in Figures 3 and 4. The HF membranes employed for NEA operate at substantially lower pressure differentials and much higher volumetric gas rates than those applied for industrial nitrogen generation. Thus they can have relatively thin walls, as can be seen in Figure 3. The perfluoropolymer layer on the outer surface of the porous polymer HF wall forms the separating layer across which oxygen permeates preferentially to nitrogen. The high flow rates for the NEA application necessitate large inside diameters and short fiber lengths in comparison to industrial membrane devices to minimize the pressure loss between the air feed and the NEA.

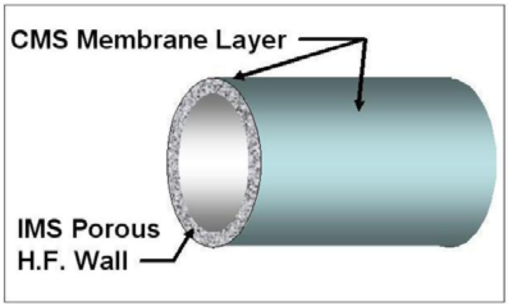


Figure 3 Hollow Fiber Membrane

Figure 4 shows a schematic of an NEA membrane device. The pressurized air feed passes along the inside of the hollow fibers that are surrounded by atmospheric pressure on their outside diameters. Since oxygen transports across the membrane more readily than does nitrogen, the feed stream becomes enriched in nitrogen and exits the device as NEA at the retentate port. The pressure differential across the HF wall provides the driving force for the dissolution and diffusion of gases across the membrane. The permeate port discharges the lower pressure, oxygen-enriched air (OEA) which is simply exhausted to the atmosphere in this application. Other requirements for the NEA membranes in the application include:

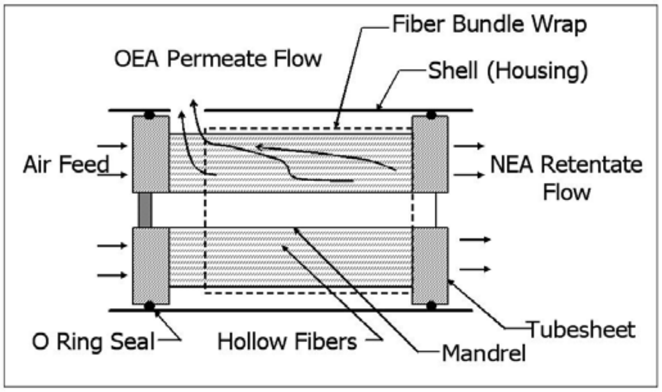


Figure 4 NEA Cartridge Cross-section

- Low pressure loss in the NEA stream;
- Moderate pressure ratios across the HF wall;
- Low loss of volume to the permeate;
- Compact size for incorporation in the intake air system;
- Good transient response;
- Robust, lightweight, and suited to engine operating environment; and
- Low manufacturing cost.

Many of these requirements are distinctly different from industrial gas membrane applications. We have already seen that the membranes for the diesel engine have distinctly different permeation properties; so too commercial membrane designs for other industrial gas separations are of little utility with diesel engines. Details of an NEA membrane installation on a diesel engine are described in the schematic in Figure 5. To meet engine application requirements, membranes need to be custom-designed to meet specific engine intake geometry and operating conditions. Membrane housings are being designed of aluminum to accommodate and protect the NEA membrane cartridge in the engine compartment.

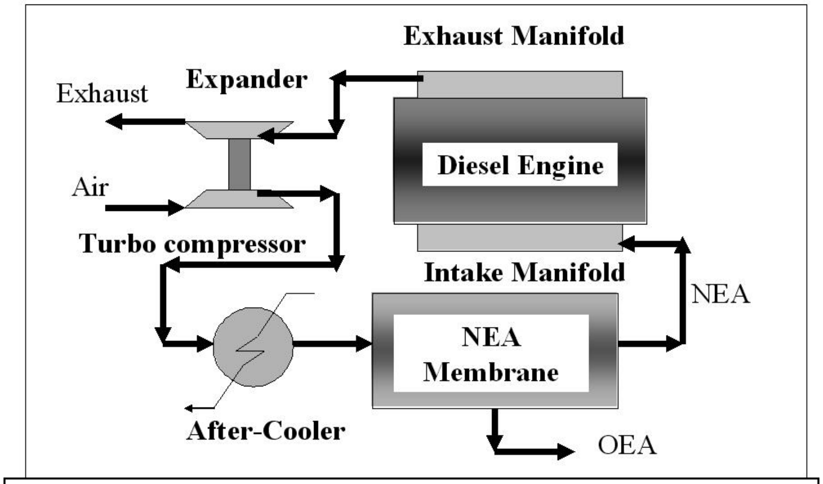


Figure 5 Flow Schematic for a Diesel Engine Outfitted with an NEA Membrane

The air charge to the diesel engine is typically pressured by a turbocharger and cooled in an aftercooler before delivery to the intake manifold. Turbochargers typically have compression ratios below 3 but this is adequate to drive the NEA membrane device as can be seen in Figure 6. Thus the NEA device can ideally be located in the air duct between the aftercooler and the intake manifold. Attention must be given to duct and membrane housing design to minimize any pressure loss between the air feed and the NEA product.

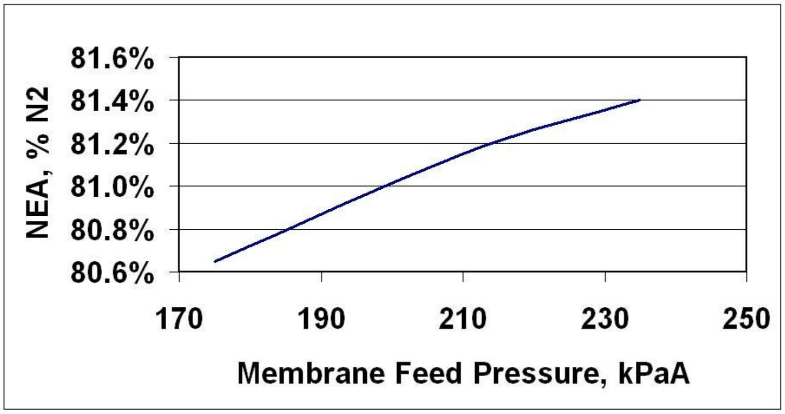


Figure 6 Typical Influence of Turbocharger Boost Pressure on Performance of Fixed Size NEA Membrane

DEVELOPMENT PROGRESS

Application of NEA membranes for NOx reduction on diesel engines has progressed from the research phase into the field demonstration stage. It is presently receiving increasing scrutiny as a viable NOx reduction option in its march toward commercialization. Thus far, four engine platforms have been employed in the evaluation and demonstration of the NEA membrane technology. The first was a 3kW Ferryman single cylinder diesel research engine. The second involved a Volkswagen 81kW 1.9 liter TDI engine, on which it was also compared against exhaust gas recycle. The third was a Lister Peter diesel electricity generator on which a variety of NEA membrane schemes were investigated. The fourth level involves recent Caterpillar C-12 diesel engine performance mapping in a test cell and NEA membrane durability evaluation on five C-12 equipped Class 8 highway trucks. These cases are summarized in Table 3.

Table 3. Four diesel engines evaluated with membrane-supplied NEA.

Description	**Driving Force**	**% NEA**	**% NOx Reduction**
A. 3 kW Laboratory Ferryman research engine	Atmos. Pressure to .3 atm vacuum	81	50
B. 81 kW 1.9L Volkswagen-Passenger Car	13 psig to atm	80	25-30
C. 20 kW 1.9L Lister-Peter LPW4T Diesel Engine Powered Generator	6 psig to .3 atm vacuum	81	to 50
D. 350 kW Caterpillar C-12 Engine in Test Cell and Highway Trucks	30 psig to atm.	81	to 50

Results of the research with the Ferryman and Volkswagen engine were reported in earlier publications [13,14,20,23,24]. It suffices to say here that the results such as those displayed in Figure 7 were sufficiently convincing as to justify continuing development of the technology.

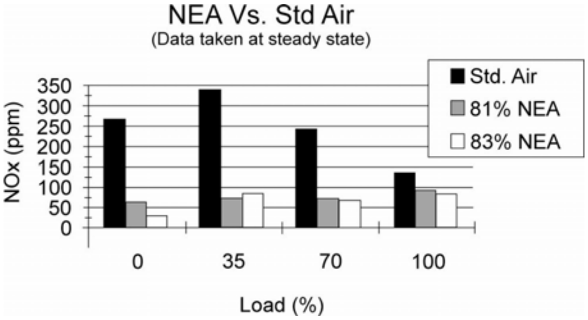


Figure 7 NOx Reduction with Nitrogen Enriched Air on Research Engine

Research on the Lister LPW4T turbocharged diesel engine was aimed primarily at investigating the feasibility of retrofitting engines with NEA to reduce their NOx emissions. Several processing configurations, including vacuum assistance to increase the driving force for the separation, were studied. As with the earlier work, it was quickly demonstrated that supplying NEA from a membrane system to the LPW4T engine substantially reduced the NOx formation. The results for only one of the process schemes investigated is reported here in Figure 8 as other schemes are the subject of intellectual property and patent applications. As with EGR, NEA addition has the effect of significant NOx reduction at the expense of some increase in the generation of diesel particulate matter. A number of approaches are being examined to minimize the formation and emission of particulates and smoke in exhausts from EGR outfitted diesel engines. Optimizing engine performance by adjusting the engine timing is a routine approach to meeting regulatory emissions limits. The addition of regenerative diesel particulate filters (DPF) on the exhaust system is also being investigated. It is expected that any of these techniques will also be compatible with the NEA scheme. Thus the modest increase in particulate emissions is not viewed as a serious impediment to either EGR or NEA schemes.

NEA was also shown to be effective in addressing NOx formation in a Caterpillar C-12 engine. An NEA system such as shown in the schematic in Figure 5 was sized for the air volume of the C-12 engine. As reported in Table 3, the turbocharger for this engine generates substantially higher boost pressures than those tested on earlier work. This is favorable to the application of the NEA membrane since the membrane area requirements are reduced with the higher pressures. The higher pressures also have the effect of more readily identifying flaws and imperfections in the membrane devices being researched. The C-12 engine program also afforded the first opportunity to operate the membrane device in a high frequency, rapid cycling, loading and unloading of the membrane devices. Fatigue points and weaknesses in the designs were quickly identified and a membrane product development effort ensued. Details of the diagnostics,

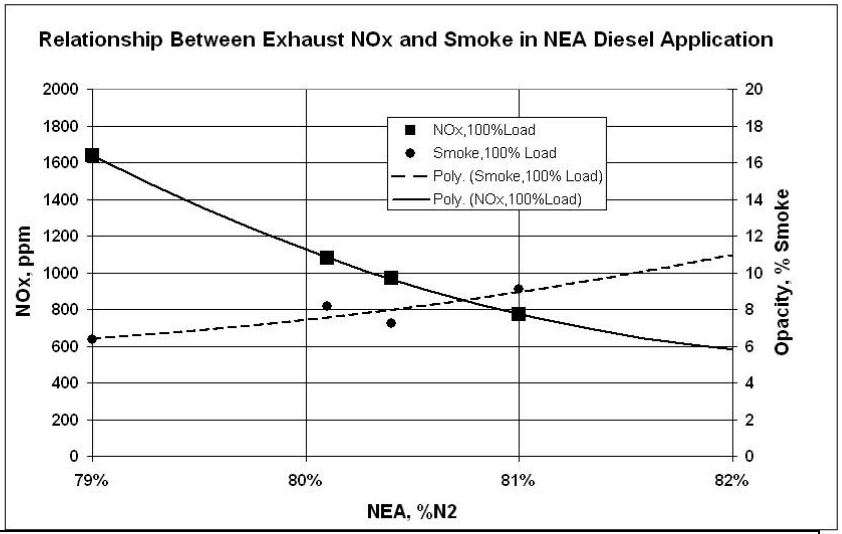
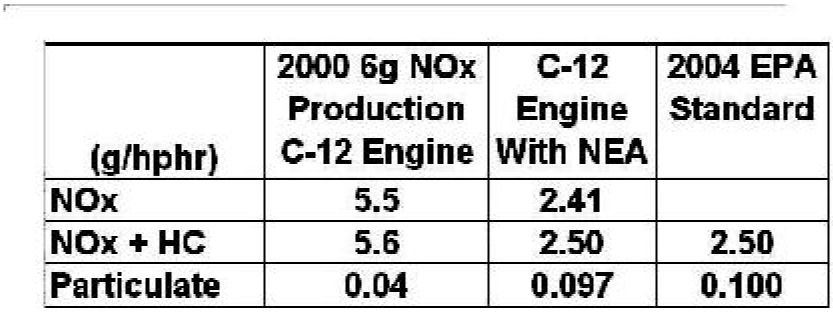


Figure 8 Effect of NEA on NOx and Smoke on the Lister Peter LPW4T Diesel Generator

Figure 9. NEA Membrane Assembly on CAT C-12 Engine on a Highway Truck.

remedies, and product durability testing involved in the perfection of the NEA membrane must go unreported for the present due to their proprietary nature. Our reporting here will be limited to the successful demonstration of the devices in the test cell and on field trials that are underway.

The volume of air required for the C-12 engine also exceeds the capacity of the largest membrane element developed by CMS and IMS for this application. This was addressed by inserting multiples of the elements described in Figure 3 in a suitable housing. The elements were arranged in a parallel fashion with headers on either end of the cartridges to insure uniform air distribution to each element while minimizing any pressure losses in the associated ducts. Figure 9 shows a photograph of the NEA membrane assembly installed between the engine and the truck hood on a highway truck. The membrane cartridges lie transverse to the top of the engine. The view in Figure 9 shows the end of the assembly where air is supplied from the aftercooler to the manifold delivering the air to the individual cartridges. NEA leaves the assembly through a manifold on the opposite end of the assembly and is ducted directly to the engine intake manifold.



(g/hphr)	2000 6g NOx Production C-12 Engine	C-12 Engine With NEA	2004 EPA Standard
NOx	5.5	2.41	
NOx + HC	5.6	2.50	2.50
Particulate	0.04	0.097	0.100

Table 5 Steady State 13 Mode Cycle Results on NEA Modified CAT C-12 Engine

A similar assembly was installed on a C-12 research engine located in an engine testing cell at the engine manufacturer. There the engine performance was measured and recorded for a series of standard tests for the steady state EPA 13 mode points. Emissions measured at these points are averaged using prescribed weighting factors to report a single statistic for rating the engine’s emissions against the EPA regulations. Table 5 provides a partial summary of the test results for NOx, unburned hydrocarbons (HC) and particulate matter (PM). As can be seen from the table, the results show the expected reduction in NOx formation at the expense of increased PM. Nevertheless, the tests demonstrate that the NEA technology is capable of meeting the 2004 EPA regulations that are being addressed with EGR and other technologies. Thus it is a NOx control technique now being considered by a number of engine manufacturers and operators for a role in their emissions reduction strategies.

As noted before, the durability of the NEA membrane devices and system is now being tested in field trials on five highway trucks outfitted with NEA membranes. All five trucks have logged over 150,000 miles each with one truck recording over 220,000 miles. There have been no indications of fouling of the membrane. One truck was pulled from the test after 151,246 miles due to low engine air flow. The membrane cartridges were retrieved and have been returned to the factory for analysis. Testing was continuing on the other four trucks.

CONCLUSIONS

On the basis of steady-state engine tests on a number of engine platforms, the following observations are made:

- NEA supplied to four widely different diesel engines with membrane devices resulted in substantial reductions in NOx emissions in all cases.
- Like EGR, NEA results in some increase in the particulate emissions.
- The NEA membrane devices can be suitably packaged and configured to operate in the engine compartment of Class 8 highway trucks.

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