DEMONSTRATION OF POTENTIAL FOR SELECTIVE CATALYTIC REDUCTION AND DIESEL PARTICULATE FILTERS

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Abstract: This project addresses the potential for Selective Catalytic Reduction (SCR) devices (using urea as reductant) together with Diesel Particulate Filters (DPF) and low-pressure loop exhaust gas recirculation (EGR) to achieve future stringent emissions standards for heavy-duty engines powering Class 8 vehicles. Two emission control systems consisting of the three technologies (EGR, SCR, and DPF) were calibrated on a Caterpillar C-12 heavy-duty diesel engine. Results of these calibrations showed good promise in meeting the 2010 heavy-duty emission standards as set forth by the Environmental Protection Agency (EPA). These two emission control systems were developed to evaluate a series of fuels that have similar formulations except for their sulfur content. Additionally, one fuel, code-named BP15, was also evaluated. This fuel was prepared by processing straight-run distillate stocks through a commercial, single stage hydrotreater employing high activity catalyst at maximum severetity.

An additional goal of this program is to provide data for an on-going EPA technology review that evaluates progress toward meeting 2007/2010 emission standards. These emissions levels were to be achieved not only on the transient test cycles but in other modes of operation such as the steady-state Euro-III style emission test known as the OICA (Organisation Internationale des Compagnies d'Automobiles) or the ESC (European Stationary Cycle). Additionally, hydrocarbon and carbon monoxide emissions standards are to be met.

Background: The APBF-DEC program is a government/industry effort addressing critical needs for emissions reduction technologies for light- and heavy-duty vehicles. It is organized into technical teams, comprised of representatives from the government and industry, to help steer the efforts that are funded by both the government and industry. Two main technical teams are addressing the most promising emissions-reducing technologies. The NO_x adsorber team (via the National Renewable Energy Laboratory) is seeking to demonstrate the potential for NO_x adsorber technology combined with diesel particulate filters and EGR to meet future emissions standards. The SCR-EGR-DPF team was tasked with demonstrating the potential for urea SCR systems together with diesel particulate filters and EGR in meeting the same objective. ORNL funded, and is administering this contract.

Other APBF-DEC technical teams provided support in fuels, urea, and lubricants and toxic emissions measurement technologies. These technical teams also included a team responsible for the experimental design and data review.

Objectives: The objectives of this project are as follows:

1. To demonstrate the low emissions performance that can be attained using low-pressure loop (LPL) EGR, urea selective catalytic reduction (SCR), and diesel particulate filter (DPF) technologies together with advanced fuel formulations for heavy-duty diesel engines. This includes regulated emissions, and selected unregulated and toxic emissions. Emissions goals are the federal 2007/2010 regulated levels for heavy-duty engines.

2. To evaluate the sensitivities of system performance parameters to fuel variables, in order to identify the critical fuel properties for emission reduction.

3. To sample for certain unregulated and toxic emissions as specified by the Technical Team. These samples will be analyzed by a third party.

4. To demonstrate the durability of the emission control systems by operating them for 6,000 hours each. The durability schedule will consist of repetitive 13-mode ESC cycles using the DECSE 8 ppm sulfur fuel.

Approach: The APBF-DEC EGR-SCR-DPF Technical Team selected a Caterpillar C-12 heavy-duty diesel engine suitable for use in this project and arranged for the engine manufacturer to provide it to SwRI. In fact a total of three engines were sent to SwRI, one engine was used for establishing the as-received performance and emission characteristics and the other two engines are to be used in long-term durability (6,000 hours on each engine). The first engine was also used to develop two control systems and their calibration toward meeting the emission targets for this program. Components of the emission control systems, such as: LPL EGR, SCR catalysts, diesel particulate filters, and urea injection systems were provided to the program by members of the Manufacturers of Emissions Control Association (MECA).

SwRI installed the development engine in a test cell capable of transient operation (Figure 1) and fully equipped for emission testing, sampling, and analysis. The engine was characterized for performance and emissions in its as-received condition. Following the initial engine characterization, it was equipped with a dual branch exhaust system. A catalyst-based DPF was installed in each branch. The dual branch exhaust was then reduced to one pipe from which EGR was provided. The engine was then equipped with the complete low-pressure loop EGR system consisting of an EGR pick up, EGR transfer pipe, EGR cooler, and a combined EGR control valve and throttle, and was calibrated to achieve reduced engine-out NO_x while maintaining good fuel economy and performance. The next step was to install two SCR catalysts with their respective urea injection system(s), and appropriate controls, and optimize the integrated system performance for maximum NO_x and PM reductions using the selected base fuel (DECSE 3 ppm sulfur). System performance was monitored as well as engine-out emissions, tailpipe emissions, regulated emissions, and selected unregulated and toxic emissions. Emission concerns that were unique to SCR systems, such as ammonia slip, were addressed as well. Two control systems, each consisting of one low-pressure loop EGR, SCR catalysts (generally one in each exhaust branch), and two catalyst-based DPFs were investigated. These systems were coded System A and System B. A schematic representation of the control system is given in Figure 2.

Figure 1. Engine Installation

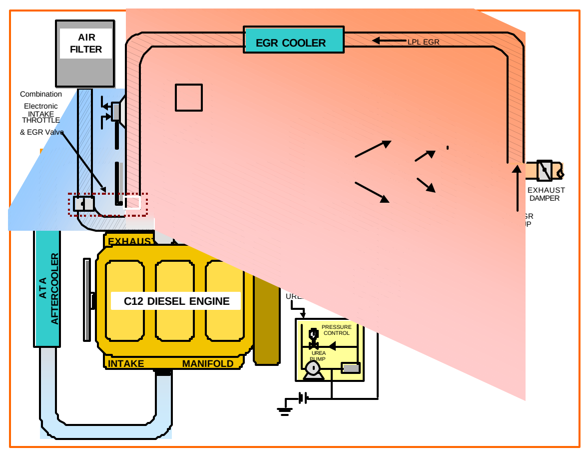

Figure 2. Schematic Representation of Emission Control System

Following the successful demonstration of system performance in the laboratory, a second phase, durability program, will be launched to age the systems over a long term (6,000 hours each) so that degradation of performance with time can be evaluated. The systems will be tested at selected intervals (2,000 hours each) for emission performance.

Fuels and Lubricants: A fuels matrix of five fuels with different sulfur levels, ranging from a nominal 3 to 30 ppm sulfur by weight, was included in the test program. These fuels were selected by the APBF-DEC team and were supplied to SwRI. The fuel formulations consisted of a baseline low-sulfur fuel, which was "doped" to higher sulfur levels except for one, the BP 15 which was specially blended at the refinery. The BP 15 was prepared by processing a straight-run distillate stocks through a commercial, single stage hydrotreater employing high activity catalyst at maximum severity. It was prepared in a commercial refinery unit but cracked stocks were excluded to achieve the low sulfur level specified. It is anticipated this process may be upgraded in the future for actual production runs. Calibrations of the EGR and urea injection schedules for the SCR catalysts were optimized at SwRI using one of the fuels (DECSE 3 ppm sulfur) as selected by ORNL and the Technical Team. After optimization on the selected fuel, SwRI ran complete tests, including emissions, with the other fuels. The durability phase of 6,000 hours on each emission control system will be conducted with one of the fuels (DECSE 8 ppm sulfur). It is important to note that the sulfur levels used in this paper are nominal and refer to original intent. The first batch of DECSE 3 ppm sulfur fuel was analyzed and the actual sulfur level was found at 4.4 ppm by weight. Another batch supplied later during System A's calibration was found to have 0.6 ppm sulfur, by weight. Fuels analysis and dopant information is given in Appendix A

The lubricant used was the same as that used in the previous DECSE program, Shell Rotella T 15W40 with sulfur content of about 3500 ppm.

Engine-out Emissions: A "2007-type" heavy-duty engine was not available for this project, and engines provided for this work were expected to produce approximately 4 g/hp-hr engine-out NO_x emissions. Details and specifications of the test engine are given in Table 1. Engine-out results when tested using DECSE fuel having 3 ppm nominal sulfur content are shown graphically in Figures 3a and 3b for transient and steady-state, respectively.

TABLE 1. CATERPILLAR ENGINE SPECIFICATIONS

Item	Description/Specification
Model Year	**2000**
Engine Model	**Caterpillar C 12**
Engine Capacity	**12.0 L/732 CID**
Power Output	**450 hp**
Rated Speed	**1800 rpm**
Peak Torque Speed	**1200 rpm**
Peak Torque	**1650 lb-ft**
Maximum Inlet Restriction	**20" H_2O**
Maximum Exhaust Pressure	**3" Hg**
Rated Speed Fuel Consumption	**0.34 lb/bhp-hr**
Injection Timing	**Variable (electronically-controlled)**
Number of Cylinders	**6**
Configuration	**In-line**
Induction System	**Turbocharged—Aftercooled**
Fuel Injection System & Control	**Unit Injector – Electronic**

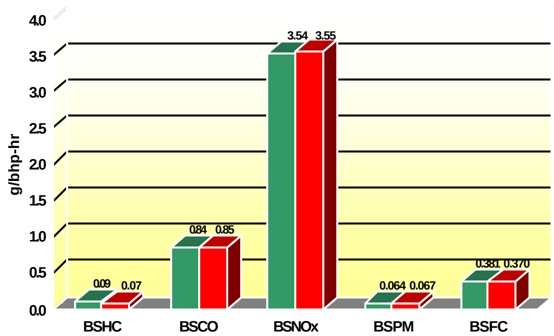

Figure 3a. Transient Results of Engine-Out Emissions and Cycle Fuel Consumption--As-Received Condition--DECSE 3 ppm Sulfur Fuel

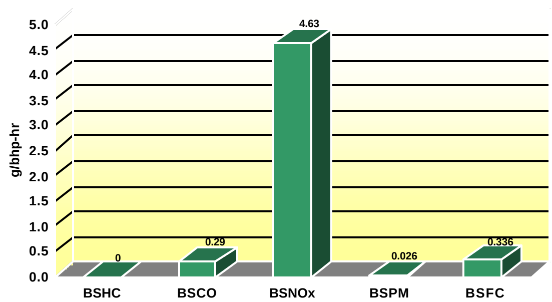

Figure 3b. Steady-State Results of Engine-Out Emissions and Cycle Fuel Consumption--As-Received Condition--DECSE 3 ppm Sulfur Fuel

Engine Emissions with Optimized EGR: A low-pressure loop EGR system was supplied to SwRI for use in reducing engine-out NO_x emissions to levels of approximately 2.0 g/hp-hr or lower. The low-pressure EGR system took exhaust from the exhaust stream after the DPF, and thus, the recirculated exhaust was mostly free of high levels of PM. SwRI was required to install the system and optimize the EGR strategy. The dual-branch exhaust system was designed, fabricated, and installed. The low-pressure loop exhaust gas recirculation system (EGR) was also installed. A photograph of the system appears in Figure 4.

A smokemeter was placed in the exhaust prior to branching into two legs as shown in Figure 4. Two catalyzed soot filters, received from the supplier of System A were installed, one in each exhaust branch also shown in Figure 4. The optimization of the system sought to minimize engine NO_x emission while preserving the engine's fuel economy characteristics. In the meantime, limits were placed on engine-out smoke, turbine inlet temperature, and compressor inlet pressure (or vacuum). Transient hot-start results are shown graphically in Figure 5, and when compared with the engine as-received condition indicate NO_x reduction with the EGR system to be about 50 percent. Similar results were obtained for the steady-state 13-mode test cycle where NO_x emissions were reduced from 4.63 to 2.32 g/bhp-hr. Emissions of HC, CO, and PM were close to zero or extremely low.

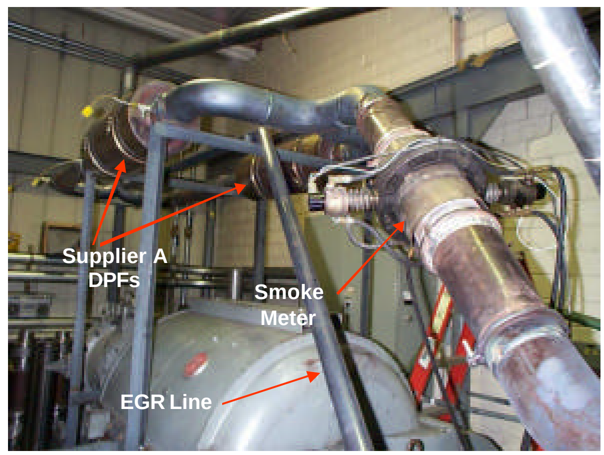

Figure 4. Dual Branch Exhaust--Smokemeter--Catalyzed Soot Filters--Low-Pressure Loop EGR Pipe

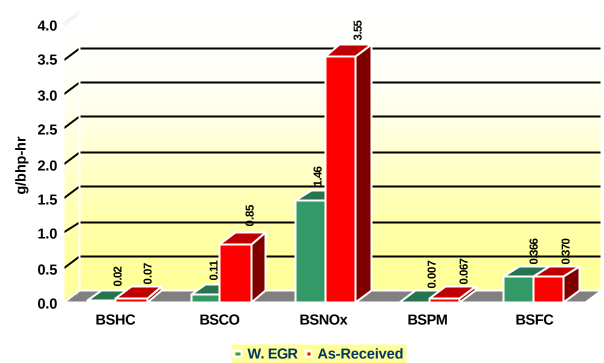

Figure 5. Hot-Start Transient Emissions and Fuel Consumption With and Without EGR

Engine Emissions With Optimized EGR, and SCR (System A): Both SCR catalysts were installed in the exhaust system as shown in Figure 6. A cleanup catalyst was also installed downstream from the SCRs, but upstream from the LPL EGR pickup. Initial calibration work was performed without the cleanup catalyst to better assess ammonia slip. An FTIR was used to monitor not only ammonia slip, but also N_2O, and NO_2. As the calibration effort progressed, it became obvious that insulating the DPFs, SCRs, and exhaust pipe was needed to improve low temperature SCR performance.

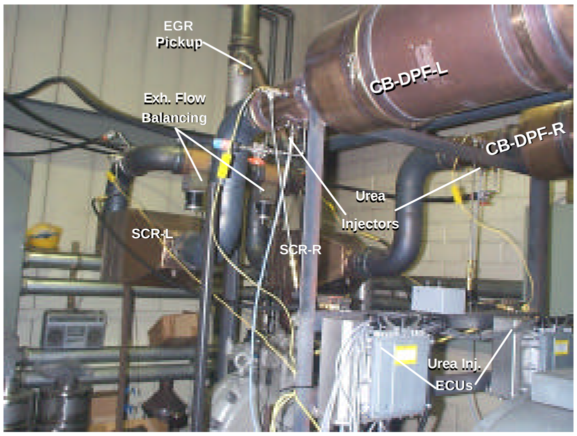

Figure 6. Exhaust System with Components of Emission Control System A--Cleanup Catalyst Not Shown

The steady-state SCR calibration was completed with satisfactory results. The OICA 13-mode composite NO_x ranged from 0.17 to 0.23 g/bhp-hr depending on ambient humidity. With low humidity, engine-out NO_x tended to be higher than with high humidity. Since the urea injection system control was basically an open-loop control system acting in response to engine speed, load, and catalyst-out temperature, ammonia slip had the tendency of increasing when humidity was high. Without closed-loop control as a function of humidity or engine-out NO_x, it will be very difficult to keep ammonia slip down without a cleanup catalyst. For the 0.17 g/bhp-hr lower limit, ammonia slip was about 3 ppm (cycle average), while the 0.23 g/bhp-hr NO_x had a corresponding 1 ppm ammonia slip. Transient results obtained with control System A are shown graphically in Figure 7.

The steady-state HC and CO results for System A with DECSE 8 ppm sulfur fuel were zero or extremely small. NO_x and PM results were 0.15 and 0.005 g/bhp-hr, respectively.

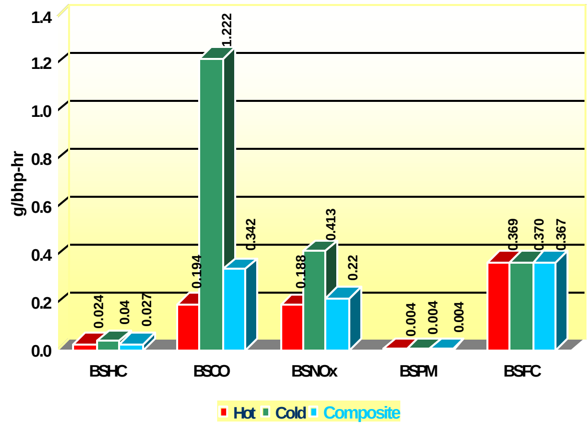

Figure 7. Emissions and Fuel Consumption Transient Results for System A - DECSE 8 ppm Sulfur

Engine Emissions With Optimized EGR, and SCR (System B): The SCR catalysts were installed in the exhaust system as shown in Figure 8. The same process used to optimize System A was once again used for System B. Due to its smaller size than System A, System B did not require insulation to improve its low temperature performance. Results of the optimized System B are shown in Figure 9

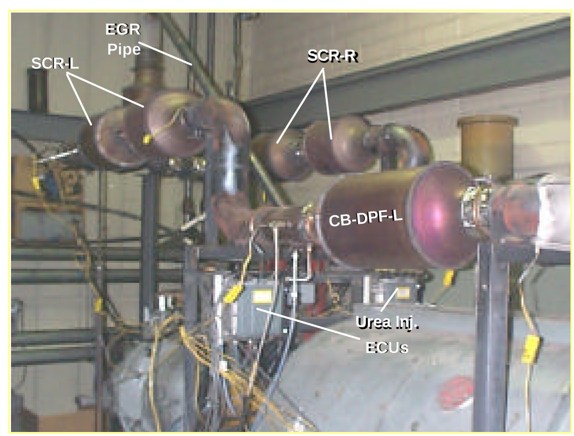

Figure 8. Exhaust System With Components of Emission Control System B

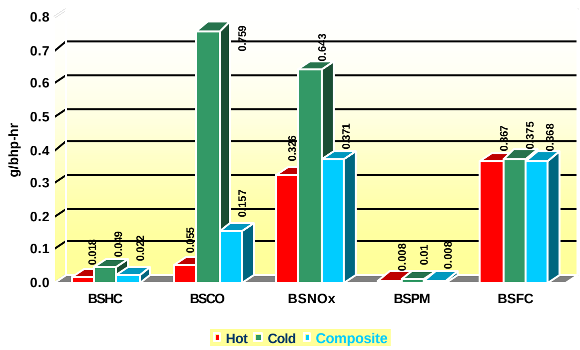

Figure 9. Transient Results of System B - DECSE 8 ppm Sulfur Fuel

Physical Comparison Between Systems A and B: Results of Systems A and B indicated a marked difference in performance. This difference is based in part on the physical size of these systems as shown in Table 2.

Emission Control Systems' Sensitivity to Fuel Sulfur: System sensitivity to fuel sulfur was assessed for both systems A and B at 10 hours following the finalization of the optimum calibration. This test was performed by purging the fuel system and connecting the supply to the test fuel prior to conditioning the engine for 4 hours at high load, alternating between peak torque and rated speeds. This procedure was followed with a practice transient cycle at the end of the day in preparation for the evaluation test the next day. A cold- followed with 3 hot-start transient tests and 2 OICA 13-mode emission test cycles were conducted. NO_x transient results for both systems at the 10-hour point and various fuels are summarized in Figure 10a and 10b and PM results for both systems are shown in Figures 11a and 11b for the transient and steady-state cycles, respectively. Conclusions regarding the effects of sulfur on emissions are pending the analysis of the larger body of data obtained at different test intervals.

TABLE 2. COMPARISON BETWEEN SYSTEMS A AND B

	No. of Units		Volume, L		Vol./Eng. Displ.		Remarks	
System	**A**	**B**	**A**	**B**	**A**	**B**	**A**	**B**
DPF	**2**	**2**	**45.6**	**34.1**	**3.8**	**2.8**	**11¼"X14"**	**10½"X12"**
SCR	**2**	**4**	**39.4**	**31**	**3.3**	**2.6**	**--**	**--**
CUC*	**1**	**1**	**8.5**	**8.5**	**0.7**	**0.7**	**--**	**--**
			93.5	**73.5**	**7.8**	**6.1**	**--**	**--**

* Cleanup Catalyst

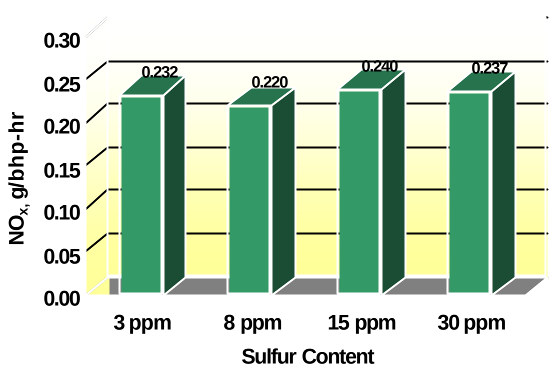

Figure 10a . Transient Cycle Composite NO_X--System A--DECSE 8 pp Sulfur Fuel

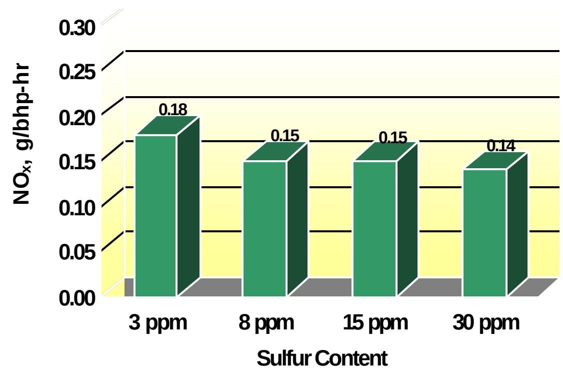

Figure 10b. Steady-State Composite NO_X--System A--DECSE 8 ppm Sulfur Fuel

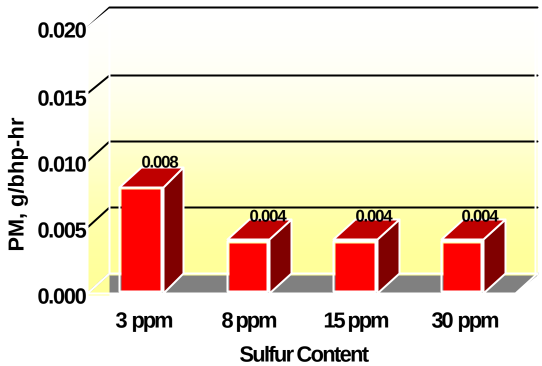

Figure 11a. Transient Cycle Composite PM--System A--DECSE 8 ppm Sulfur Fuel

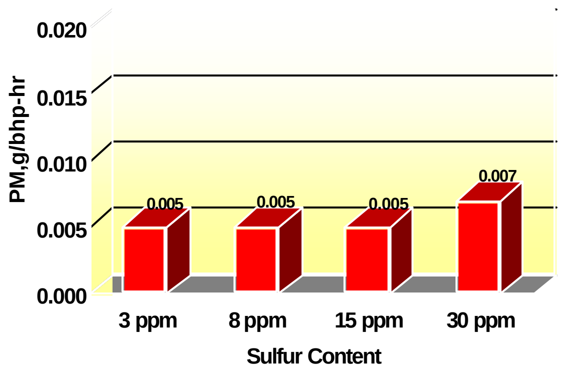

Figure 11b. Steady-State Composite PM—System A--DECSE 8 ppm Sulfur Fuel

Future Work: A planned 10-,100-, and 200-hour fuels evaluation will be completed using both Systems A and B. This evaluation will be carried out according to the same test protocol established for this program, namely by performing one cold-start followed by 3 hot-start EPA transient tests and two 13-mode OICA steady-state tests for each fuel.

Following the 200-hour fuels evaluation, Phase 2 will begin and continue for 6,000 hours on each of the two emission control systems (A and B). This phase will consist of three 2,000-hour segments. At the end of each 2,000-hour segment, a complete evaluation, using the same protocol, will be conducted.

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APPENDIX A

FUELS ANAYSIS

Fuel Property	ASTM Test	DEC Value	DEC Goal	BP15 Value
Density, kg/m3	D4052	826.2	820-850	837.1
Viscosity @ 40C, mm2/s	D445	2.3	>2.0	2.5
Distillation IBP, C	D86	180	171-182	164
10% recovery, C	D86	203	210-226	201
20% recovery, C	D86	219		218
30% recovery, C	D86	233		233
40% recovery, C	D86	244		246
50% recovery, C	D86	251	254-271	259
60% recovery, C	D86	257		272
70% recovery, C	D86	265		286
80% recovery, C	D86	279		302
90% recovery, C	D86	312	310-321	322
FBP, C	D86	352	326-360	346
Cloud point, C	D2500	-26		-12
Pour point, C	D97	-23		-18
Flash point, PMCC, C	D93	69	>52	64
Sulfur, ppm	D5453	0.6	<10	13.3
Aromatics, vol. %	D1319	23.9	25-32	29
Olefins, vol %	D1319	4.6	1-3	
Saturates, vol. %	D1319	71.4	55-70	
Aromatics, wt. %	D5186	26.9		25
Polyaromatics, wt. %	D5186	8.4	3-10	4.2
Non-aromatics, wt. %	D5186	64.7		70.8
Cetane number	D613	42.5	42-48	51.1
Cetane index	D976	51.5		48.8

DOPANT DETAILS

Amount, Mass %	Constituent	Formula	Boiling Point, C
50	Dibenzo[b,d]thiophene	$C_{12}H_8S$	333
30	Benzo[b]thiophene	C_8H_6S	222
10	Di-t-butyl disulfide	$C_8H_{18}S_2$	200
10	Ethyl phenyl sulfide	$CH_{10}S$	206

- A mixture of four sulfur-containing compounds
- Rationale
  - Represent a variety of classes of sulfur-containing compounds: sulfide, disulfide, thiophene and dibenzothiophene
  - All compounds in diesel boiling range
  - Emphasize dibenzothiophenes (50% of mixture)
  - Commercial available
- Dopant was added gravimetrically
- Sulfur level of base fuel determined though determinations by multiple laboratories