Effect of oil sands processing severity on the combustion and emissions of an HCCI engine

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Canada

Effect of Oil Sands Processing Severity on the Combustion and Emissions of an HCCI Engine

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1. Introduction

Canadian oil sands derived diesel fuels normally contain a larger fraction of aromatics and cycloparaffins than conventional crude-derived diesel fuels. The hydrogenation process used in bitumen upgrading converts polynuclear aromatic molecules to monoaromatics and saturates. It also typically converts some of the high boiling point fractions to lower boiling point components, as well as increases the cetane number. The process is complex and requires significant hydrogen. The result is a premium synthetic crude oil that is suitable for further processing by refineries into diesel fuels that meets the standards specified for use in conventional diesel engines.

Homogeneous charge compression ignition (HCCI) is a potentially promising combustion strategy because it offers the potential for diesel-like efficiency with near-zero NO_x and soot emissions. The question asked is whether HCCI combustion requires the same level of hydrotreating as used to produce current diesel fuels in the marketplace.

The hydrotreating process hydrogenates aromatic hydrocarbons, first converting polynuclear aromatic compounds (containing multiple aromatic rings) into mono-aromatics and saturates [1]. While a modest improvement of diesel fuel quality is possible by converting polynuclear aromatics to monoaromatics, further increases in cetane number require extensive hydrotreating to first saturate the monoaromatics, [2] and then open the rings to form paraffins.

The relationship between fuel cetane number (CN) and HCCI combustion characteristics is still not fully understood. A few studies have highlighted the relationship between cetane number and fuel properties. For example, in a series of studies by Yui [1-5], the relation between cetane number, sulphur, aromatics, hydrogen, aniline point, and mid boiling point were examined for a matrix of diesel fuels undergoing hydrotreating. It was shown that the hydrotreating process increased the hydrogen content of the fuel, while the aromatic content, sulphur content, and mid boiling temperature were all decreased. It was shown that cetane number was improved by increasing hydrotreating severity. The relation between cetane number and aromatic content was more complex. Based on experimental results, three different trends were observed. For hydrotreated, fluid coker, light gas oils, cetane number decreased as aromatics increased. For the rest of the gas oils, cetane number increased as aromatics increased, and for the wide boiling range components, cetane number increased whereas aromatics increased and decreased [1]. However the generally accepted trend is that cetane number increases when aromatics is decreased.

The current study compares the performance of three oil sands derived diesel fuels, subjected to different hydrotreating severities, in an HCCI engine. The study is focused on the relationship between fuel chemistry

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variation (caused by hydrotreating) and HCCI combustion characteristics. Several researchers have investigated fuel chemistry effects on HCCI combustion timing for gasoline and diesel fuels. Aroonsrisopon et al. [6] examined fuels with identical research octane numbers, but different fuel chemistries. They found that HCCI combustion is a strong function of fuel composition and cannot be predicted by octane number. In a series of studies by Shibata el al. [7-10], the strong influence of fuel composition on HCCI combustion characteristics was emphasized. Especially for fuels exhibiting dual-stage autoignition behaviour, it was shown that fuel chemistry directly affects low temperature heat release characteristics, and the subsequent main combustion stage. It was found that aromatics, olefins, and some cycloparaffins such as cyclopentane reduce LTHR activity. Examining a matrix of 11 fuels, a new fuel quality index was developed for HCCI combustion based on LTHR and LTHR inhibition effects such as those of aromatics [9]. This may be the most promising method of rating diesel fuels for HCCI combustion as it considers the importance of LTHR and its relation to cetane number and other fuel characteristics.

In a series of studies by Bunting et al. [11, 12], the effect of cetane number on HCCI combustion was examined. It was found that high cetane fuels have stronger LTHR behaviour and require lower intake temperature for autoignition. Statistical analyses of the experimental results for a range of diesel fuels showed that fuel efficiency was mainly controlled by fuel properties such as density and energy content and that the autoignition characteristics were related mainly to cetane number.

2. Experimental Setup

A single-cylinder, four-stroke, variable compression ratio, Cooperative Fuels Research (CFR) engine was used for this study. The engine was coupled to an eddy current dynamometer for measuring engine load and controlling engine speed. A variable speed AC motor, coupled to an over-drive clutch, was used to motor the engine prior to initiating HCCI combustion. The engine setup was modified from the standard CFR configuration by the addition of an air-assist port fuel injection system and hardware to control critical engine parameters such as intake temperature, air/fuel ratio, intake and exhaust pressures, and exhaust gas recirculation.

A port fuel injector, designed for flexible fuel vehicles, was used to atomize the fuels just upstream of the intake port. Blast air was supplied to improve the atomization process. The blast air was taken from the measured intake air after the mass flow meter. For this study, the fuel system and assist-air pressures were maintained at 500 kPa and 200 kPa, respectively. The timing and duration of the air blast relative to the fuel injection event was optimized to minimize emissions. Experimental data from this study shows that this fuel injector produces droplets with an SMD (Sauter Mean Diameter) less than 15 micrometers. An in-house fuel vaporizer was added downstream of the fuel injector to vaporize the lighter fractions of the diesel fuel, which enables a more homogeneous air-fuel mixture to form inside the combustion chamber as evidenced by near-zero soot emissions. The fuel vaporizer temperature (T_{vap}) was measured at the end of heated section of the vaporizer.

The intake air was dry, pressurized air that was metered and supplied to the intake surge tank. The air was mixed with recycled exhaust gases inside the intake surge tank. The intake air and EGR mixture then passed through a lengthy pipe section which provided additional time for the mixture to homogenize, thereby avoiding large temporal variations in EGR fraction. One of the elements was a heat exchanger to cool the air and EGR mixture. The heat exchanger coolant was heated to just below the intake temperature set point to avoid water vapor condensation inside the heat exchanger. The heat exchanger was sized appropriately to compensate for the small temperature difference. A second heater located upstream of the fuel vaporizer was used to supply heat to the air/EGR mixture (T_{air}) to achieve the desired intake mixture temperature (T_{mix}) measured in the intake manifold downstream of the fuel vaporizer.

The engine exhaust was passed through an exhaust surge tank. A back pressure exhaust valve was used to maintain the exhaust pressure slightly above the intake pressure to provide the pressure drop to recycle the exhaust gases to the intake stream.

All experiments were performed under steady state conditions. An engine data acquisition and control system (Sakor Technologies Inc., DynoLab™) was used to acquire engine temperatures, pressures, and flows. Exhaust gas concentrations (NO_x, HC, CO, CO_2, O_2) were measured using emissions analyzers (California Analytical

Instruments). An Artium Technologies' Laser Induced Incandescence (LII) instrument measured exhaust soot concentration. A water-cooled Kistler pressure transducer (Kistler Corp., model 6041A) was flush-mounted in the cylinder head. The cylinder pressure data were acquired for 300 consecutive engine cycles using an AVL IndiModule combustion analyzer with 0.2 CAD resolution. Further heat release analyses and indicated calculations were performed by in-house MATLAB® software.

3. Fuels

In this study, a straight-run light gas oil (a relatively low cetane quality refinery stream) derived from oil sands sources was subjected to two different levels of hydrotreating severity. The resultant three oil sands derived fuels and an ultra-low sulphur diesel (ULSD) fuel derived from conventional petroleum were evaluated in an HCCI engine. The first fuel (OS-CN35) was straight-run light gas oil with a 35 cetane number. This low-cetane fuel was upgraded using the PP13 hydrotreating unit at the National Centre for Upgrading Technology (NCUT) to obtain two additional fuels with cetane numbers of 38 and 42. The two hydrotreated fuels had lower densities and viscosities as well as higher H/C ratios. The more deeply hydrotreated fuel with the higher cetane number had a significantly reduced distillation profile compared to the straight-run light gas oil. The hydrocarbon composition analyses show that progressive hydrotreating transformed the unsaturated aromatics into mono- & di-cycloparaffins, and then into iso- & normal-paraffins. Table 1 summarizes properties of the fuels examined in this study.

Table 1: Properties of examined fuel matrix

	ASTM Method	OS-CN35	OS-CN38	OS-CN42	ULSD
Cetane Number	D613	35	38	42	47
Carbon, wt%	D5291M	86.48	86.08	85.28	86.22
Hydrogen, wt%	D5291M	13.34	13.87	14.44	13.70
Sulphur, mg/L	D5623	54	38	8	<15*
Density, g/mL	D4052	0.8351	0.8248	0.7859	0.8189
Viscosity, cSt @ 40°C	D445	1.633	1.531	1.109	1.465
Sim. Distillation, T10, °C	D2887	158	150	119	165
Sim. Distillation, T50, °C	D2887	229	222	190	216
Sim. Distillation, T90, °C	D2887	291	286	255	273
Saturates	PIONA & GC-FIMS	72.4	81.9	96.2	77.6
i- + n-paraffins	PIONA & GC-FIMS	26.1	27.7	47.0	41.6
Monocycloparaffins	PIONA & GC-FIMS	20.4	24.3	43.9	20.3
Dicycloparaffins	PIONA & GC-FIMS	17.0	21.4	4.3	11.4
Polycycloparaffins	PIONA & GC-FIMS	8.9	8.5	1.0	4.3
Total Aromatics	PIONA & GC-FIMS	27.5	17.9	3.4	22.0
Monoaromatics	PIONA & GC-FIMS	26.0	17.7	3.4	20.2
Diaromatics	PIONA & GC-FIMS	1.5	0.2	<0.1	1.8

*estimated

4. Results and Discussion

Minimum Vaporizer Temperature

The vaporizer temperature (T_{vap}) was adjusted for each fuel while maintaining a fixed intake mixture temperature to determine how much external fuel vaporization was required to achieve near-zero soot emissions. Figure 1 shows that a different critical fuel vaporizer temperature was needed to achieve low soot emissions for each fuel. The critical temperatures were found to be 210°C, 200°C, and 160°C for the 35, 38 and 42 cetane number fuels, respectively. The vaporizer temperature was set to 20°C higher than the critical temperature for subsequent experiments.
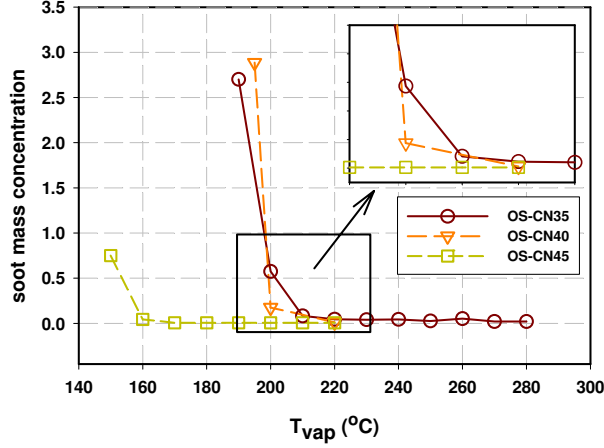
Figure 1: Effect of fuel vaporizer temperature on soot emissions measured by Artium laser-induced incandescence. All other operating conditions (T_{mix}, AFR, EGR, N, CR) were kept constant for each fuel.

Note that T_{vap} was varied in 10°C increments. Values obtained for the critical temperature from Figure 1 are well correlated with distillation temperatures indicated in Table 1. For soot-free combustion, the fuel needs to be completely vaporized before combustion starts. While the fuel is partially vaporized in the fuel vaporizer, the higher boiling point molecules vaporize during the compression stroke. Thus, the T90 temperature provides important information about the critical temperature required to achieve a completely homogeneous mixture.

Fixed Speed-Load Conditions

The effect of hydrotreating severity on HCCI combustion and emissions was investigated at nine speed-load conditions, as summarized in Table 2. In general, it was assumed that each fuel would require a different combination of operating (CR) and initial conditions (T_{mix}, AFR, and EGR) in order to achieve optimal HCCI combustion.

Thus, different initial and operating conditions were permitted in order to satisfy the performance objectives of high thermal efficiency, near-zero NO_x and soot emissions, reasonable HC and CO emissions, and limited rates of pressure rise and combustion noise. For the fuels investigated in this study, it was possible to operate the engine up to a compression ratio of 13.0 for all fuels. Mode 1 is the limiting condition for compression ratio because the engine speed is low, while IMEP and MAP are high. Mode 1 has the maximum low temperature heat release and hence requires the lowest intake heating, lowest compression ratio or highest EGR fraction.

Table 2: Fixed speed-load operating conditions

IMEP (bar)	900 (rpm)	1250 (rpm)	1600 (rpm)	MAP (bar)
6 →	Mode 1	Mode 4	Mode 7	← 2.0
4 →	Mode 2	Mode 5	Mode 8	← 1.5
2 →	Mode 3	Mode 6	Mode 9	← 1.0

The strategy was to fix the mixture temperature at the minimum possible value for each fuel, unless there was a necessity to advance combustion timing, subject to the following two restrictions: no water condensation in the EGR line; and near-zero soot emissions. AFR and EGR were adjusted for each fuel and at each mode to achieve peak rates of pressure rise below 10 bar/°CA and acceptable combustion cyclic variations ($COV_{IMEP} < 5\%$). Also, for 6 bar IMEP, EGR was adjusted to achieve stoichiometric AFR, and for 2 bar IMEP operating condition, EGR was maximized, then AFR was maximized until $COV_{IMEP} = 5\%$ was reached. If 2 bar IMEP was not reached, EGR was reduced gradually. HC and CO emissions were monitored under idling conditions and if excessive further EGR reductions were made.

The intake mixture temperature (T_{mix}) was maintained at 75°C for all fuels. CA50 was fixed at TDC for all fuels and modes, except for modes 3, 6, and 9 where the combustion timing was retarded with zero EGR and the maximum possible AFR. The combustion timing could have been advanced by increasing the intake mixture temperature or compression ratio, but the intention was to keep them constant for all fuels. Figure 2 shows the indicated specific fuel consumption for all fuels at the nine modes. The experimental data shows that OS-CN42 exhibited the best efficiency, followed by OS-CN38 and OS-CN35.
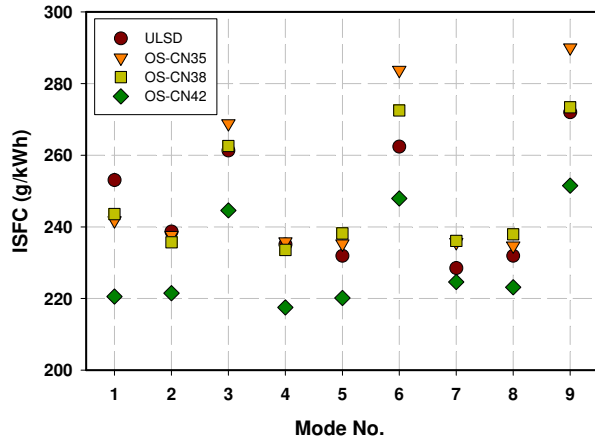
Figure 2: Indicated specific fuel consumption for fixed speed-load conditions of Table 2.
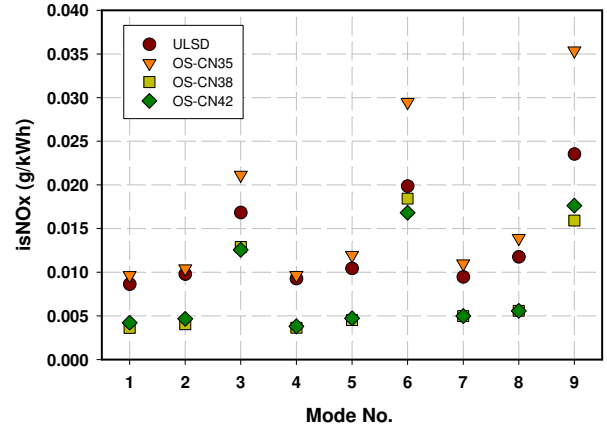
Figure 3: Indicated specific NO$_x$ emissions for fixed speed-load conditions of Table 2.

In particular, OS-CN42 had the lowest ISFC at modes 3, 6, and 9 which all had retarded combustion timings and high cyclic variations. Note that combustion timing, speed, load, and initial conditions are fixed at each mode, hence the observed trends of decreased ISFC with increasing hydrotreating severity was due to fuel chemistry differences. As several fuel properties were changed by hydrotreating, it is difficult to relate the decreased ISFC to a specific property in Table 1. However, looking at combustion characteristics shows that despite maintaining constant combustion timing, the combustion duration is reduced with increasing hydrotreating, rates of pressure rise are increased, and combustion noise (quantified by knocking index) is increased (not shown). This may well be related to the selective ring opening that occurred for the severely hydrotreated fuel OS-CN42. Faster combustion increases efficiency as it approaches ideal Otto cycle constant volume combustion. Also, reduced HC and CO emissions lead to higher combustion and thermal efficiencies.

Figure 3 shows the two hydrotreated fuels derived from oil sands sources, OS-CN38 and OS-CN42, had the lowest NO$_x$ emissions. These two fuels had the highest hydrogen contents and the lowest aromatics. Under similar combustion timing conditions, it is expected that at the same power and speed, peak combustion temperature is relatively constant. Increasing hydrotreating severity decreased NOx emissions due to the shorter combustion duration that reduced the time available for NOx formation.

At these extremely low temperatures, NOx is not formed by conventional thermal mechanisms. It is more likely formed by HCN and prompt mechanisms, so small peak combustion temperature increases by hydrotreating does not have an adverse effect on NOx emissions. The prompt mechanism is attributed to the reaction of atmospheric nitrogen, N$_2$, with radicals such as C, CH, and CH$_2$ fragments derived from the fuel, where this cannot be explained by either the aforementioned thermal or fuel processes. Occurring in the earliest stage of combustion, this results in the formation of fixed species of nitrogen such as NH (<u>nitrogen monohydride</u>), HCN (<u>hydrogen cyanide</u>), H$_2$CN (<u>dihydrogen cyanide</u>) and CN- (<u>cyano</u> radical) which can oxidize to NO.

Note that decreasing load for each constant speed line (for example from mode 1 to 3) increased NOx due to reduced efficiency. NOx concentration (ppm) however was decreased. Figures 4 and 5 show the modal HC and CO emissions, respectively. Consistent with the ISFC data, modes 3, 6, and 9 produced very high HC and CO emissions for all fuels. The lowest cetane fuel, OS-CN35, produced the highest HC emissions, particularly at the three low IMEP modes. Test fuel OS-CN42, on the other hand, generally produced the lowest HC and CO emissions at all modes. This is consistent with the observation of reduced combustion duration and the hypothesis of increased peak combustion temperatures.
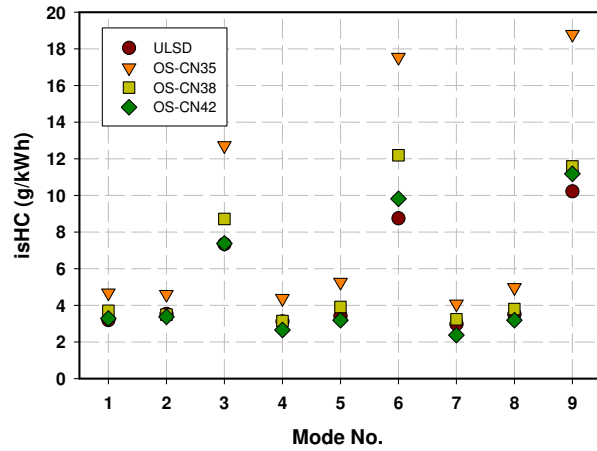
Figure 4: Indicated specific HC emissions for fixed speed-load conditions of Table 2.
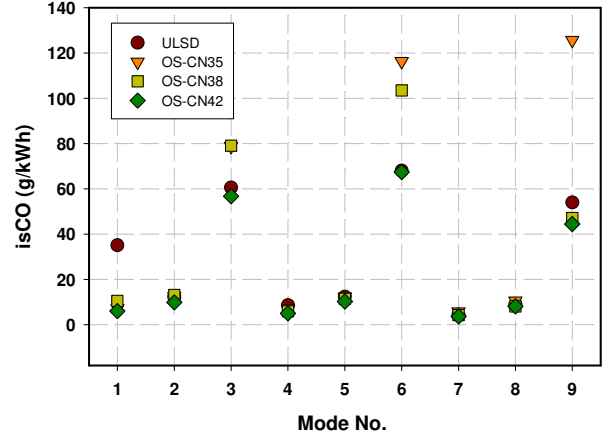

Figure 5: Indicated specific CO emissions for fixed speed-load conditions of Table 2.

Fixed Initial Conditions

In the previous section, the speed-load conditions were fixed, while the initial and operating conditions were allowed to vary for different fuels. Another approach is to examine the range of initial conditions (AFR, EGR) over which HCCI combustion was achievable without fixing the engine power, speed, and/or combustion timing.

The AFR –EGR operating region is limited on the rich side (low AFR, low EGR) by knocking. The criteria for the knock limit was set at $(dP/d\theta)_{max}$=10 bar/^{o}CA. The operating region is limited on the lean side (high AFR, high EGR) by misfiring and/or partial burning. The criteria for the lean limit was set at COV_{IMEP}=5%. Hence, all operating points indicated in Figure 6 correspond to $(dP/d\theta)_{max}$≤10 bar/°CA and COV_{IMEP}≤5%. Note that the expansion of experiments toward higher AFR and lower EGR fractions and was not done due to the fact that optimal HCCI combustion in terms of power, efficiency, and emissions does not occur in this region.

Figure 6 shows the attainable EGR-AFR operating region for the three oil sands derived fuels and the commercial ULSD fuel. The knocking boundary corresponds to the high power limit at a given EGR rate and is an indication of high rates of pressure rise, faster combustion, and shorter ignition delay. As hydrotreating severity was increased from OS-CN35 to OS-CN38 to OS-CN42, eventually both the rich and lean limits were shifted toward leaner mixtures, and the width of the operating window between the rich and lean limits was expanded. Limitation of the rich limit by increased hydrotreating level causes a reduction in the maximum power for certain operating conditions. By increasing hydrotreating severity, HCCI combustion became faster and hence the knocking tendency was increased. The rates of pressure rise were higher for the highest cetane oil sands fuel (OS-42CN) than for the lowest cetane fuel (OS-CN35). Consequently, the rich side of the operating window was shifted toward leaner mixtures. This will impose a problem in a practical HCCI combustion engine as it limits the maximum power. The general statement that low cetane number fuels are better for HCCI combustion comes from this phenomenon. The same reasoning can be used to explain the expansion of the lean limit. The highest fuel conversion efficiency occurs on the rich side of the operating region near the knocking boundary. Expanding the lean limit does provide the potential advantage of load control at constant EGR rate by adjusting AFR over a wider range.

5. Conclusions

The effects of processing severity on the HCCI combustion characteristics and emissions were examined experimentally for three Canadian oil sands derived diesel fuels in a single-cylinder, variable compression ratio, HCCI engine. The three fuels were a light gas oil fuel (OS-CN35), the product of a mild hydrotreating process (OS-CN38), and the product of a fairly severe hydrotreating process (OS-CN42). The hydrotreating process generally leads to increased cetane number, decreased aromatic content, and often a change in the distillation profile towards lighter fractions. Two types of experiments were performed. The first type was output-driven, in which a series of

fixed speed-load conditions were examined for all fuels. The second type was input-driven, in which the initial conditions were changed without controlling engine speed, load, or efficiency.
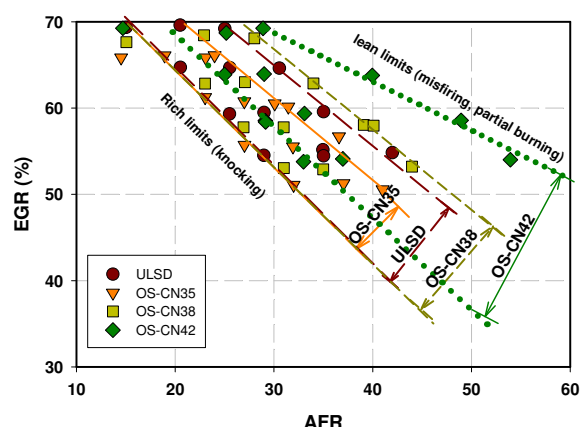

Figure 6 : Comparison of operating region for all fuels, CR=13.0, N=900 rpm, T_{mix}=75°C, T_{vap}=220°C

It was found that the fuel vaporizer temperature required for near-zero soot emissions decreased with increasing hydrotreating severity due to increased fuel volatility.

At fixed speed-load and combustion timing, more severe hydrotreating reduced the combustion duration, and increased rates of pressure rise that resulted in improved efficiency and emissions. Based on HC and CO emission trends, it was concluded that combustion temperature was slightly higher for the hydrotreated fuels. Despite the slightly higher temperatures, NOx emissions were reduced due to the shorter available time for NOx formation.

The hydrotreating process expanded the AFR-EGR operating window of the oil sands fuels and shifted the operating region toward leaner mixtures.

This is a possible disadvantage for heavily hydrotreated fuels as it limits the maximum power.

Nomenclature

AFR	Air/fuel ratio
COV_{IMEP}	Coefficient of variation in indicated mean effective pressure
CR	Compression ratio
EGR	Exhaust gas recirculation
IMEP	Indicated mean effective pressure (bar)
LTHR	Low temperature heat release (J)
MAP	Manifold absolute pressure (bar)
N	Engine speed (rpm)
T_{mix}	Intake mixture (air, EGR, fuel) temperature (°C)
T_{vap}	Vaporizer outlet temperature (°C)

References

1. S. Yui, *Oil & Gas Journal* **2000**, 58-66.
2. S. Yui, *Oil & Gas Journal* **2005**, 46-51.
3. S. Yui, *Prepr. Pap.-Am. Chem. Soc., Div. Pet. Chem.* **2006,** 51, (1&2).
4. S. Yui, *Hydrocarbon Processing* **2007,** July 2007 99-105.
5. S. Yui; K. H. Chung, *Oil & Gas Journal* **2007**, 52-59.
6. T. Aroonsrisopon; D. Foster; T. Morikawa; M. Iida, *SAE Paper* **2002,** 2002-01-1924.
7. G. Shibata; K. Oyama; T. Urushihara; T. Nakano, *SAE Paper* **2004,** 2004-01-0553.
8. G. Shibata; K. Oyama; T. Urushihara; T. Nakano, *SAE Paper* **2005,** 2005-01-0138.
9. G. Shibata; T. Urushihara, *SAE Paper* **2007,** 2007-01-0220.
10. G. Shibata; T. Urushihara, *SAE Paper* **2008,** 2008-01-0007.
11. B. Bunting; C. Wildman; J. Szybist; S. Lewis; J. Storey, *International Journal of Engine Research* **2007,** 8, (1), 15-27.
12. B. G. Bunting; R. W. Crawford; L. R. Wolf; Y. Xu, *SAE Paper* **2007,** 2007-01-4059.