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The Emerging Market for Biodiesel and the Role of Fuel Additives

P Richards, J Reid
Innospec Limited

L-H Tok
Innospec Limited (Singapore)

I MacMillan
Innospec Fuel Specialties LLC

ABSTRACT

With growing concern over greenhouse gases there is increasing emphasis on reducing CO_2 emissions. Despite engine efficiency improvements plus increased dieselisation of the fleet, increasing vehicle numbers results in increasing CO_2 emissions. To reverse this trend the fuel source must be changed to renewable fuels which are CO_2 neutral.

A common route towards this goal is to substitute diesel fuel with esterified seed oils, collectively known as Fatty Acid Methyl Esters. However a fundamental change to the fuel chemistry produces new challenges in ensuring compatibility between fuel and engine performance/durability.

This paper discusses the global situation and shows how fuel additives can overcome the challenges presented by the use of biodiesel.

INTRODUCTION

Towards the end of the nineteenth century a German engineer considered the existing internal combustion engines to be wasteful of fuel and patented a system wherein the fuel was added during the expansion stroke at a controlled rate such that all the fuel energy was converted to work and no excess heat was produced (1). This he believed would significantly increase the overall efficiency of the engine. Whether this ideal was ever achieved is open to question but by virtue of reduced pumping loses and higher compression ratio the compression ignition engine, now commonly known as the Diesel engine after this German engineer, has traditionally exhibited higher efficiency than its spark ignited competitor. However with the increasing availability of fuel and its concomitant reduction in relative price the emphasis in engine development has been towards increased power density, which generally meant higher engine operating speeds. This was more readily achieved by pre-mixing the fuel and air and hence the spark ignition engine became dominant, especially for smaller engine size applications.

In the second half of the twentieth century the increasing popularity of the internal combustion engine began to cause concern in particular geographic locations due to the amount of air pollution that these engines were creating. Thus was born emissions control legislation. With the introduction of this legislation, fuel producers and engine manufacturers have been working to reduce the emission of the regulated pollutants. However step reductions in emissions have in cases resulted in a step increase in fuel consumption. With the exception of the Corporate Average Fuel Economy (CAFE) rules in the USA, fuel consumption was not legislated. In Europe and Asia fiscal pressure has been the driver for improved fuel economy. This has undoubtedly been a factor in the increasing dieselisation of the European passenger car fleet. In the trucking industry price has always been an overriding consideration throughout the globe. This has thus favoured the diesel engine for these applications. In the emerging economies of India and China there is thus increasing demand not only for diesel fuel per se but for diesel fuel as a percentage of the fuel slate.

Whilst there is still pressure to reduce the currently regulated pollutants, attention is being focused on the global effects of vehicle emissions. With growing concern over climate change and the role of greenhouse gas (GHG) emissions there is increasing emphasis on reducing CO_2 emissions. Despite the improved efficiency of both gasoline and diesel engines plus the increased dieselisation of the fleet, the increased size of the vehicle fleet has resulted in increasing CO_2 emissions.

Figure 1 shows project light vehicle production by geographic region until the year 2012 (2). This figure shows that light vehicle production in Greater China is projected to double by 2010 with global production up by more than 27% by the year 2012.

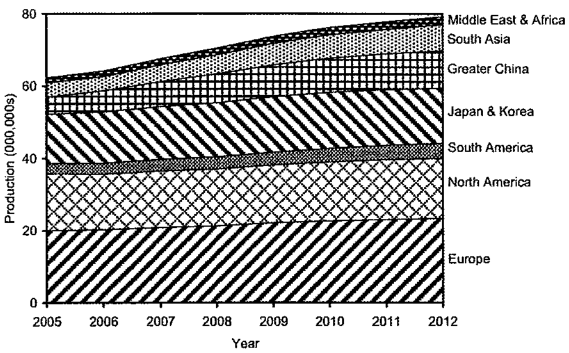

Figure 1. Projected light vehicle production

It has therefore been concluded that to slow, halt or ultimately reverse the increase in CO_2 emissions the source of fuel energy must be changed to renewable fuels which are CO_2 neutral.

The use of plants as a raw material for fuel production is seen by many as an answer to this problem. There is currently much work ongoing to determine the true benefit of using this approach once the energy cost of fertilising, harvesting, transporting and processing the crop has been considered (3-12).

Of the many proposed options for converting plant material to fuel the one that is currently most common is the esterification of seed oils to produce what are collectively known as Fatty Acid Methyl Ester (FAME), which can be used as a substitute for diesel fuel, when it is commonly referred to as B100 or as fuel blending component for diesel fuel when the finished fuel is commonly referred to according to the FAME content by volume, for example B5 for a fuel containing 5% volume of FAME or B20 for a fuel containing 20% volume of FAME. However a fundamental change to the chemistry of the fuel produces new challenges in ensuring compatibility between fuel and engine performance and durability.

The use of FAME as a diesel fuel extender or substitute is not new, the first national standard for the specification and testing of a FAME was introduced in Austria in 1991 (13). This covered RME as a diesel fuel substitute. Five years later this was widened to cover FAMEs in general (13). Other European countries introduced there own national standards and by 2003 there was a European Union wide standard (EN14214) covering FAME. At around the same time the Australian Biodiesel Standard was introduced along with ASTM D6751 in the USA. These standards are very similar, the US standard having a slightly lower cetane number limit and a higher sulphur limit. At the time of writing a Japanese specification was at the draft stage, this was essentially the same as EN14214 but with a tighter limit on Total Acid Number (TAN).

FAME can also be derived from animal fats and reclaimed vegetable or animal derived fats such as used cooking oil. The FAME used as biodiesel consist of a range of components that are traditionally classified according to the number of carbon atoms in the molecule and the number of double bonds, which is a measure of the degree of saturation of the acid from which the FAME is derived. So for example a fully saturated compound with 18 carbon atoms (Stearic acid) would be C18:0 whilst the unsaturated equivalent (Oleic acid) would be C18:1 and the polyunsaturated versions Linoleic and Linolenic acids would be C18:2 and C18:3 respectively. A list of the acid equivalents of the FAMEs typically found in biodiesel is given in Table 1.

Table 1. FAMEs typical found in biodiesel

Fatty Acid		Structure
Caprylic	C8:0	$CH_3(CH_2)_6COOH$
Capric	C10:0	$CH_3(CH_2)_8COOH$
Lauric	C12:0	$CH_3(CH_2)_{10}COOH$
Myristic	C14:0	$CH_3(CH_2)_{12}COOH$
Palmitic	C16:0	$CH_3(CH_2)_{14}COOH$
Palmitoleic	C16:1	$CH_3(CH_2)_5CH{=}CH(CH_2)_7COOH$
Stearic	C18:0	$CH_3(CH_2)_{16}COOH$
Oleic	C18:1	$CH_3(CH_2)_7CH{=}CH(CH_2)_7COOH$
Linoleic	C18:2	$CH_3(CH_2)_4CH{=}CHCH_2CH{=}CH(CH_2)_7COOH$
Linolenic	C18:3	$CH_3(CH_2)_2CH{=}(CHCH_2CH{=})_2CH(CH_2)_7COOH$
Arachidic	C20:0	$CH_3(CH_2)_{18}COOH$
Eicosenoic	C20:1	$CH_3(CH_2)_7CH{=}CH(CH_2)_9COOH$
Behenic	C22:0	$CH_3(CH_2)_{20}COOH$
Eurcic	C22:1	$CH_3(CH_2)_7CH{=}CH(CH_2)_{11}COOH$

In the same way as the chemical composition of petroleum derived diesel fuel is dependant upon the crude oil from which it is derived, the chemical composition of FAMEs is dependant upon the original vegetable source and this can produce a significant variation as shown in Figure 2. This figure shows the composition of FAMEs derived from different sources; coconut oil methyl ester (CME), palm oil methyl ester (PME), rape oil methyl ester (RME), soy oil methyl ester (SME), tallow methyl ester (TME) and reclaimed used cooking oil methyl ester (UCOME).

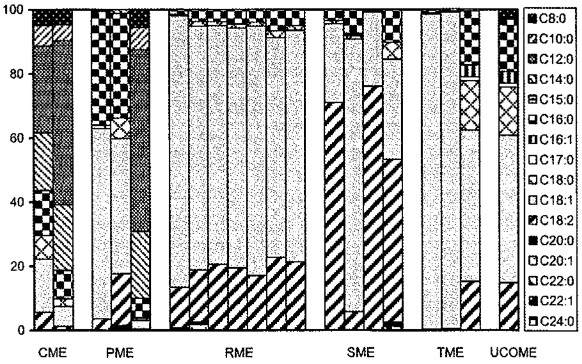

Figure 2. Composition of different FAMEs

With the exception of the UCOME the chart shows the composition of more than one example of the different FAMEs, this shows that there can be significant differences in the composition of what is ostensibly the same product, although looking at the chart it could be postulated that some of the samples have been misrepresented. This in itself is a point of concern but is outside the scope of this paper. The way in which these different compounds affect different fuel properties will be discussed in later sections of the paper.

The quality of the FAME is not only influenced by the source of the fatty acid but also by the esterification process, in the same way as petroleum diesel is not only influenced by the crude oil source but also the oil refinery process. Ideally all impurities are removed from the fatty acid and all the acid is converted to the corresponding ester, however in practice this does not happen and other impurities are introduced. The European standard therefore specifies that the biodiesel (B100) must contain a minimum of 96.5% FAME. So for example the biodiesel may contain un-reacted fatty acid and un-reacted methanol, water, glycerides, glycerol, metals either from the esterification process or from the source material plus sulphur from the source material.

There is therefore the potential for significant variation in product quality and specification for products generically described as FAME. This has led to numerous initiatives by Original Equipment Manufacturers (OEMs) (15) and their Industry Associations (16) to publicise this issue and the potential for FAME degradation in the fuel supply chain and vehicle fuel systems. Their studies have shown that these reactions (which are accelerated by the presence of oxygen, water, heat and certain impurities) can result in the formation of corrosive materials (such as formic, acetic and organic acids, water and methanol) and polymers that may drop out of solution when mixed with fossil derived diesel.

The following vehicle operability problems have been identified during extensive field trials as being caused by these fuel characteristics:-

- Low temperature operability
- Compatibility with Fuel Injector Equipment (FIE) components
- Elastomer compatibility
- Deposit formation
- Increased dilution and polymerisation of engine sump oil
- Tailpipe emissions

How diesel fuel additive packages must be tailored to address these concerns is discussed in the following sections.

LOW TEMPERATURE OPERABILITY

The ability of the fuel to flow at low temperature is obviously of prime importance. It must also continue to flow over time. Some fuels will form small wax crystals as the temperature falls, whilst these crystals do not prevent the fuel from flowing they will collect in the fuel filter and will rapidly clog the filter thus preventing any further fuel flow. It is believed that a high level of saturated compound in the FAME will result in poorer cold flow properties (17).

The CFPP of a combination of FAME and diesel fuel is difficult to predict. This is clearly shown in Figure 3. This shows the CFPP of four different base diesel fuels (labelled as D1 to D4) when blended with 5% and 20% RME and 5% and 20% SME. The base diesel fuels had a CFPP of between -4°C and -34°C while the neat RME and SME had a CFPP of -18°C and -3°C respectively.

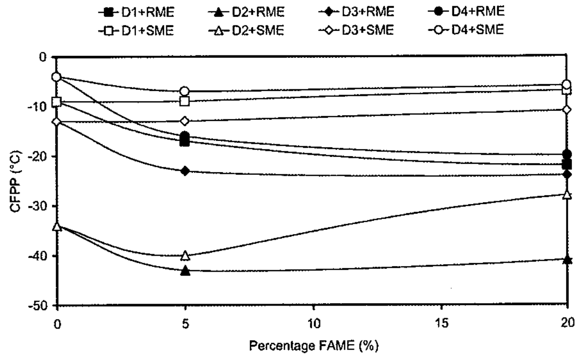

Figure 3 CFPP of blended fuels

Three of the base fuels have a higher CFPP than the RME but with the addition of only 20% RME the CFPP of the blend is ostensibly the same as that of the RME. It is also interesting to not from this chart that both the RME and the SME which have a far higher CFPP than the diesel fuel D2 both depress the CFPP of the base diesel fuel when blended at low concentrations of FAME.

The response of FAME to cold flow improvers (CFI) is also difficult to predict. Figure 4 shows the reduction in CFPP as a result of using two commercial CFIs on a number of FAME samples. As can be seen from the chart CFI "B" significantly reduces the CFPP of the RME samples but has little effect on the CFPP of the other FAMEs. CFI "A" on the other hand has a similar effect to CFI "B" on two of the RME samples but a noticeably lower effect on the remaining three RME samples. Again there is little effect on the other FAME samples. Both CFIs were used at the same high treat rate.

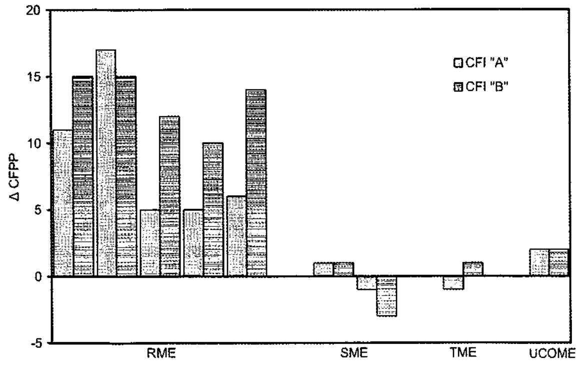

Figure 4. Change in CFPP with addition of CFI

This suggests that it is difficult to lower the CFPP of SME, however blends of diesel and SME can be treated effectively, although as the concentration of SME in the blend increases then the benefit from the CFI decreases. This is illustrated in Figure 5 which shows the reduction in CFPP brought about by the use of two commercial CFIs, "G" and "H", in B5 and B20 blends of SME in a base diesel that had a CFPP of -13°C. The CFPP of the SME used was -5°C.

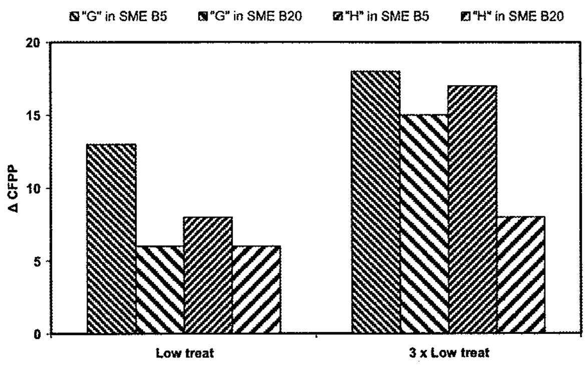

Figure 5 Effect of CFIs on SME blends.

This figure also shows the effect of CFI treat rate on the change in CFPP. From this it can be seen that CFI "H" responds well in the B5, reducing the CFPP by 8°C at the low treat rate and 17°C at the triple treat rate, however it has a poor response in the B20, reducing the CFPP by 6°C at the low treat rate but only 8°C at the triple treat rate. On the other hand CFI "G" although it has a greater effect than CFI "H", it does not have as good a response in the B5 but has a good response in the B20. CFI "G" is therefore clearly more suitable for SME blends.

FIE COMPATIBILITY

The fuel injection equipment has always been a major influence on the performance of a diesel engine. To meet ever more stringent emissions regulations while still meeting the performance demands of the user, including fuel economy, the diesel fuel injection equipment has become increasingly sophisticated and ever more precisely engineered. These high precision pieces of equipment are designed to be compatible with prevailing fuels without being over-engineered, this ensures a cost effective piece of equipment. A change in the fuel quality not only produces a challenge in designing new equipment it also causes concern as to how the new fuel will react with existing equipment that may have been in use for many years.

The concerns of FIE manufacturers are discussed below. Free methanol from the production process can cause corrosion of aluminium and zinc components; it will also lower the flash point which is of safety concern. Unreacted fatty acids can also cause corrosion of non-ferrous metals. Incomplete washing and filtering of the FAME can also leave inorganics within the FAME that can lead to filter plugging or increased wear in the fuel pump and injection nozzles. Free water can cause corrosion and increases the risk of bacterial growth which may cause filter plugging. Free water can also promote hydrolysis of the FAME which then reverts to fatty acid and methanol which can lead to the problems mentioned above. Free glycerine and glycerides can produce lacquering on pumps and injectors. The FAME itself may cause softening, or hardening, swelling or cracking of different elastomers which may result in fuel leakage. The FAME may displace deposits from the diesel resulting in filter plugging.

Even when these concerns are overcome for the fresh FAME or FAME blends the effects of aging are still of concern. As FAME ages it can produce formic and acetic acid which will corrode the metal parts in the FIE. It can also polymerise, leading to deposits which may block filters or cause lacquering and deposit formation on the fuel injectors.

Regarding oxidation stability, the European standard EN 14214 specifies a minimum induction period of 6 hours measured according to the test method EN 14112. The induction period is the period of time before aged FAME produces a measurable amount of volatile acids. The FAME is aged at 110°C under a constant stream of air, this air carries away the volatile acids formed in the FAME, these acids are then adsorbed in demineralised water, the conductivity of which acts as the measure of acids generated in the FAME.

In a recent survey conducted in the USA (18) only one out of twenty-eight samples tested had an induction period of greater than 6 hours. In a Japanese study none of the FAME samples had an induction period of greater than 6 hours (19).

This problem can again be readily countered by the use of a carefully selected additive. Figure 6 shows the effect of three stability additives on RME, SME and a 50/50 blend of RME and SME.

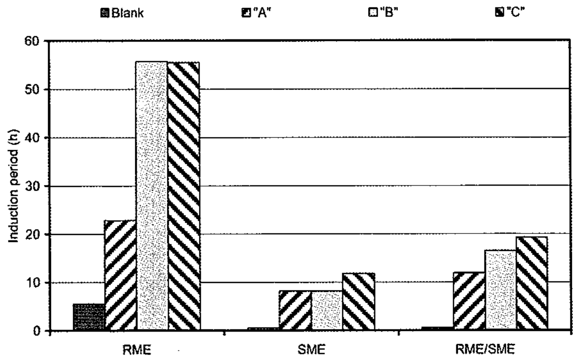

Figure 6 Effect of stability additives on induction period

It is clear that additives "B" and "C" are far more effective than additive "A" in the RME whereas only additive "C" shows superior performance in the SME. The RME/SME blend shows the expected relative ranking of the three additives although the magnitude of the effect appears to be dominated by the SME component. The aforementioned Japanese study (19) also considered the use of anti-oxidants to increase the oxidation stability of the FAMEs tested.

In most cases the anti-oxidant did increase the oxidation stability but with two samples of RME the use of the antioxidant increased the level of total insolubles. The exact reason for this was unclear but it was postulated that the level of water, carbon residue, methanol and potassium in these samples was a contributory factor.

It is thus clear that with the correct choice of stability additive the FAME induction period can be extended well beyond the limit of EN 14214. The importance of doing so was demonstrated in a recent study to assess the impact of biodiesel blends on FIE durability (20). This study assessed B20 blends using RME and SME that had been treated with a commercially available antioxidant and a B20 blend using untreated SME that had been oxidised by sparging with clean dry air with the FAME maintained at 57°C. The study included comparison of the different B20 blends using an industry standard rotary pump test (CEC F-32-X-99). The test with the highly oxidised SME B20 failed to reach the end of the test due to fuel filter blocking. At this point the test fuel had separated into two distinct phases suggesting that the fuel had undergone decomposition during testing. It must be noted however that this fuel did not indicate increased pump wear up to this point. This is discussed further below.

Another point of concern is the effect of fuel system metals on the stability of biodiesel blends. Copper is well known for de-stabilising diesel fuel. Copper will also destabilise biodiesel when present in even small quantities but because of the effect with petroleum diesel fuel the incidence of copper in diesel fuel systems is low, however a recent report also found that die-cat aluminium can also accelerate the degradation of biodiesel fuels (21).

WEAR

It is widely accepted the FAME has inherently good lubricity (22). The inclusion of FAME into ultra low sulphur diesel fuel can compensate for the loss of lubricity brought about by the hydro-treatment employed as part of the de-suphurisation process (23). However one study (24) did conclude that "Contaminants in biodiesel, especially free fatty acids and monoacyglycerols are largely responsible for the lubricity of low level blends (such as 1-2%) of biodiesel with low-lubricity petrodiesel." A study considering B5 and B20 blends of RME and SME and including an oxidised SME B20 blend assessed the wear on a conventional rotary diesel pump, a high pressure common rail pump and pintle type injectors (21). The conclusions of this work were that the B5 and B20 blends made with the FAME samples containing an anti-oxidant did not increase wear beyond that of the conventional diesel fuel. Tests with the heavily oxidised SME B20 blend did not reach a conclusion due to the poor stability of this fuel leading to filter plugging.

ELASTOMER COMPATIBILITY

When a hydrocarbon fuel comes into contact with an elastomer there is a tendency for the elastomer to absorb some of the hydrocarbon, there is also the possibility that the fuel will act as a solvent; removing some of the soluble components from the elastomer. The former mechanism usually predominates and the net result is swelling of the elastomer. The degree of swelling is determined by a combination of the properties of the elastomer, the cross-link density, the amount and type of filler, etc (25), and the properties of the fuel; its chemical composition, density, viscosity, polarity and temperature. Polar compounds have been found to cause significant swelling of fluorosilicone and nitrile butadiene rubber (NBR) (26).

However a more recent study (27) comparing petroleum diesel fuel and B100 found no difference in the swelling, elongation, hardness or tensile strength of flourosilicone rubber. A further study which included a B20 blend made from highly oxidised SME (21) concluded that fluorocarbon elastomers with medium to high fluorine content are most compatible with the B5 and B20 fuels tested and that other candidate elastomers showed good resistance to change in physical properties but exceeded the typically acceptable levels of degradation in one or more tests.

DEPOSIT FORMATION

Due to the fact that FAME is more susceptible to oxidation than petroleum diesel fuel and that degraded biodiesel is known to produce gums and lacquers the use of biodiesel has caused concerns over increased levels of deposit formation. To reduce engine-out emissions FIE manufacturers have developed fuel injectors with smaller injection holes relying on higher fuel pressure to ensure a more finely atomised fuel spray. This technology is hence more susceptible to deposit formation.

At the date of writing there is no agreed test method for evaluating deposit formation in the latest common-rail fuel injection systems. Evidence from older technology engines is often conflicting. A paper investigating the injector tip deposits for engines powering transport refrigeration units (TRUs) (28) showed that B100 lead to hard black deposits that interfered with the optimum spray pattern of the injectors within about 1000 hours of operation.

A further study (29) which relied on the strip-down of vehicles, that had been operating in service for a number of years using B20 concluded that there was little difference in operational and maintenance costs that could be attributed to the fuel. However the vehicles that had been operated on the B20 fuel had a higher frequency of injector replacement.

As part of an in-house study a heavy duty truck that had been running on neat petroleum diesel fuel, conforming to EN 590, was switched to a commercial RME B100 fuel. Prior to the fuel change a fuel injector was removed from the engine, which had covered over 200,000 km. The injector was examined using a scanning electron microscope (SEM). This showed a small but acceptable level of deposit formation in the fuel injection holes. An example of this deposit formation is shown in Figure 7.

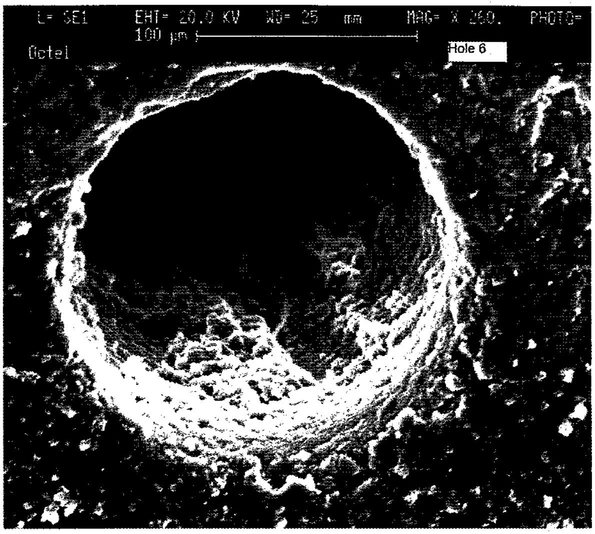

Figure 7. Injector deposits running on diesel fuel

The engine was then switched to run on B100 and the vehicle was run for a further 60,000 km, an injector removed at this point an subject to SEM examination showed a much lower level of deposit formation. This is shown in Figure 8.

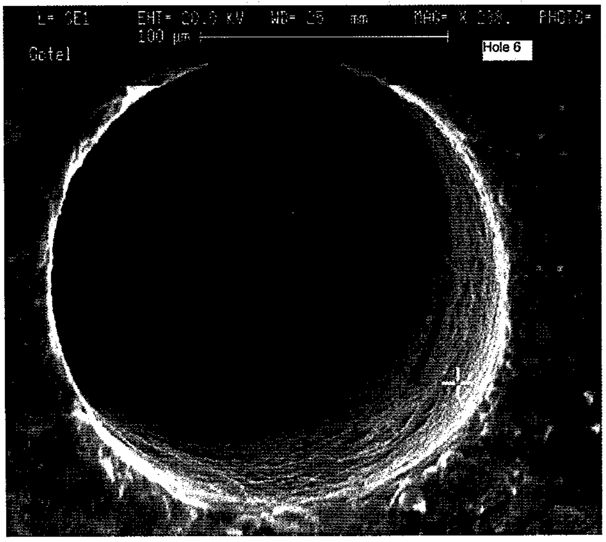

Figure 8. Injector deposits running on B100

ENGINE EMISSIONS

The use of FAME as a blending component in diesel fuel is widely recognised as significantly reducing the emissions of hydrocarbons (HC), carbon monoxide (CO) and particulate matter (PM). This can be attributed to the oxygen content of the FAME, unfortunately this also contributes to a lower calorific value for the FAME blended fuel which will reduce the maximum power of engines run on such blends. There are however isolated studies that show an increase in PM emissions (30).

Some researchers have however reported an increase in CO emissions (31). It is interesting to note that this study also observed "Only the storage life of the biodiesel is a parameter which needs to be studied further as a negative variance with regards to emissions performance was observed with biodiesel under long durations of storage. This is a well known feature of biodiesel which can be remedied by using appropriate additives."

The effect of blending FAME into diesel fuel on NO_X emissions is less clear. Whilst some researchers have found that FAME tends to increase NO_X emissions (32, 33) others have found that FAME decreases NO_X emissions (34-36). It has also been reported that the use of di-tertiary butyl peroxide (DTBP) and 2-ethyl-hexyl-nitrate (2EHN) can negate the increase in NO_X observed with some B20 blends (37). DTBP and 2EHN are two of a large number of compounds that are known to increase the cetane number of petroleum derived diesel fuels; it is also well know that increasing the cetane number of diesel fuel tends to reduce NO_X emissions. Recent work (38) has tried to isolate the possible mechanisms by which FAME may affect the NO_X emissions from an engine and concluded "Cetane number can be an inaccurate measure of ignition delay for biodiesel relative to PRF blends at operating conditions typical of modern diesel engines." The same study also indicated that with biodiesel reduced soot radiative heat transfer and mixture strength at the lift-off length may play a role in increasing NO_X emissions; however these two parameters are related.

The effect of biodiesel on unregulated emissions is obviously not as extensively studied. However some studies that have been carried out indicate that further investigation is important if the request to reduce CO_2 emissions does not result in the increase in some other unregulated pollutant.

A German study (39) concluded that the use of RME leads to and increase in ozone precursors, the emissions of aldehydes and alkenes were indicated as being responsible for this. A later Canadian study however found that a B20 using used cooking oil reduced ozone forming potential by 43% (40).

A Greek study (30) also found significantly higher acetaldehyde emissions with a slight upward trend for formaldehyde emissions.

CONCLUSION

The use of FAME as a substitute for petroleum diesel is already a reality, primarily as a blend of up to 5% FAME in petroleum diesel. The use of blends of up to B20 is commonly proposed. There are also cases of the use of B100. The main driver for this is the reduction in life-cycle CO_2 emissions. The use of FAME can also have a beneficial effect on the regulated emissions of species, however further work is required to ensure that non-regulated pollutant emissions are not increased.

Two of the major obstacles to widespread acceptance of higher FAME use are stability of the FAME and FAME blends plus the low temperature operability issues. Work presented here along with previously published work shows that these problems can be overcome with good quality control and the use of appropriate fuel additives.

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CONTACT

For further information contact

Paul Richards Tel: +44 151 350 6914
Fax: +44 151 356 6112
Email: paul.richards@Innospecinc.com

Jacqueline Reid Tel: +44 151 350 6913
Fax: +44 151 356 6112
Email: jacqueline.reid@innospecinc.com

Innospec Limited
Innospec Manufacturing Park
Oil Sites Road
Ellesmere Port
Cheshire
CH65 4EY
England

Lim Hoe Tok Tel: +65 6830 8504
Fax: +65 6336 1692
Email: LimHoe.Tok@sg.innospecinc.com

Innospec Limited
Branch Office Singapore
47 Scotts Road
#02-03 Goldbell Towers
Singapore 228233

Ian MacMillan Tel: +1 (302) 4511357
Fax: +1 (302)
Email: Ian.MacMillan@lab.innospecinc.com

Innospec Fuel Specialties LLC
200 Executive Drive
Newark
Delaware 19702
USA

http://www.innospecinc.com