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Aldehydes Emissions Measurement and OFP assessment of Biodiesel and its Blends with Kerosene using a Low NOx Gas Turbine Combustor

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ABSTRACT

There is more interest of using biodiesel fuels derived from vegetable oil or animal fats as alternative fuels for both diesel and gas turbine engines. This is mainly due to the potential benefits on CO_2 reductions and renewable. Regulated emissions of biodiesel and its blends are widely studied in diesel engines and some gas turbine engines. However, there is a knowledge gap of lack of information about non-regulated pollutants such as carbonyl compounds (aldehydes etc). This paper assessed aldehydes emissions under atmospheric pressure and 600K using a radial swirler industrial low NOx gas turbine combustor. A comparison was made between B100 (100% WME), B20 (80% Kerosene: 20% WME) and pure Kerosene. A FTIR was used to determine aldehydes including formaldehyde, acetaldehyde and acrolein. OFP (Ozone Formation Potential) of formaldehyde emissions were assessed for these three fuels. The results showed that formaldehyde was the most prevalent aldehyde species for B100, B20 and kerosene, accounted for up to 50%. The aldehydes decreased as equivalence ratio increased due to the increased flame temperatures. A strong correlation between aldehydes emissions and flame temperatures was observed.

NOMENCLATURE

CBCs: carbonyl compounds.

CO: carbon monoxide.

EI: Emission Index.

FTIR: Fourier Transform Infrared.

OFP: Ozone Formation Potential.

PM: Particulate Matter.

THC: Total unburned Hydrocarbon.

WME: Waste cooking oil Methyl Ester.

INTRODUCTION

Negative environmental consequences of fossil fuels and concerns about petroleum supplies have spurred the search for alternative fuels [1, 2]. In the last decade, biodiesels have received significant attention whether it was obtained from methyl/ethyl esterification of fatty acids from vegetable oils, animal fats or used frying oils [1]. Biodiesel can be produced by transesterification process of vegetable oils or animal fats with alcohol in the presence of catalyst to achieve a viscosity close petrodiesel. It has similar physical properties with petroleum derived diesel and can be mixed with diesel [3]. Biodiesel and its blends with diesel have been tested in diesel engines without modifications by many researchers and the most common blending ratio is 20% of biodiesel, termed B20 [2, 4]. The main conclusion is that with biodiesel fuel there was a significant reduction in major emissions (UHC, CO, PM) with the exception of NOx emissions [5, 6]. However, the use of biofuels in the transport and power generation is a highly controversial area. On the positive side, these appear to offer a means of reducing net greenhouse gas emissions within the capabilities of current engine designs. Conversely, there are ethical concerns over the use of food crops and concerns over the ability of advanced non-food biofuels to be produced on the scale required to have a significant impact on transport and power generation is a highly controversial. Hence, there is a clear need for a fully informed cost-benefit analysis of biofuels, including all aspects of impacts.

There are much limited studies investigated the unregulated emissions from biodiesel such as carbonyl compounds (CBCs) especially aldehydes. These compounds are playing an important role in atmospheric chemistry and urban air. Also, they are a source of major health concern and important precursors to free radical ozone [7]. It is reported that formaldehyde, acetaldehyde acrolein and methyl ethyl ketone are the main species of carbonyl emissions from the exhaust and are toxic, mutagenic and even carcinogenic to human body [1, 8].

Traffic emissions are one of the most important sources of formaldehyde and acetaldehyde pollutions in urban air. The formation of them is a result of incomplete combustion of fuel. There are two pathways of formation of formaldehyde and acetaldehyde in urban air: primary and secondary. The primary source is emitted directly from the vehicles through exhaust gases. The secondary sources are formed from photo oxidation of other VOCs emitted from exhaust. Formaldehyde is the most dominant aldehyde in vehicle exhaust emissions. Both formaldehyde and acetaldehyde are classified as a probable human carcinogen by EPA in 1987 and exposure to them can cause irritation of the eyes, nose and respiratory tract.

The relationship between engine conditions and formaldehyde emissions has been studied in a diesel engine and the results showed that at lowest load conditions formaldehyde emissions was more than twice as high as that at the higher load conditions[5]. Lin et al investigated the emissions of carbonyl compounds from a heavy duty diesel engine using five different biodiesels. The study summarised that formaldehyde was the major carbonyl in the exhaust accounting for 70.3~75% of the total CBC concentration for all fuels. The effect of aldehydes emissions with equivalence ratio in a light duty diesel engine was studied by Zervas et al. They concluded that aldehydes decrease as the equivalence ratio increases and they represent from 8 to 16% of the pollutants detected [5].

There is a knowledge gap in aldehydes emissions from gas turbines, especially formaldehyde. Li et al [7] investigated unregulated emissions from a micro gas turbine engine using the FTIR. They found that the major detected aldehydes (formaldehyde, acetaldehyde and acrolein) had similar level of emissions at low power (<20% of maximum thrust) and at maximum power (>60% of the thrust), formaldehyde emissions increased rapidly. Formaldehyde has a strong potential to form ozone and is classified as an active ozone formation precursor. Ozone is irritant gas that can pose hazards to mucous membranes of eyes and respiratory tract,

The work in this paper compared the emissions between a waste cooking oil derived methyl ester (WME) and kerosene using a radial swirler industrial low NOx gas turbine combustor under atmospheric pressure and 600K preheated air. The pure WME (B100) and its blends with kerosene (B20) and pure kerosene were tested for formaldehyde, acetaldehyde and acrolein emissions. The OFP was assessed.

EXPERIMENTAL TECHNIQUES

Combustion Rig

The premixed gas turbine combustor test rig was used. The apparatus used for the experiments consisted of a combustion chamber, an air supply line, a venture air flow meter, a 250mm diameter air plenum chamber, a fuel supply line and an exhaust gas analyze system as shown in figure 1. The combustion experiments were carried out at atmospheric pressure. The air was supplied by a pump and heated to the required inlet temperature (600K) by 150KW electrical heaters. A 6.4mm thick stainless steel uncooled combustor, cylindrical shape with an inner diameter of 140mm and length of 330mm was used in this study. A 76mm outlet diameter radial double swirler was used to stabilize the flame and improve the mixing as shown in figures 2 and 3. A 45º eight flat bladed co-rotating radial double swirler with 25mm hub diameter was used to house a central fuel injection. The two swirlers were separated by a splitter plate with 40mm diameter outlet orifice.

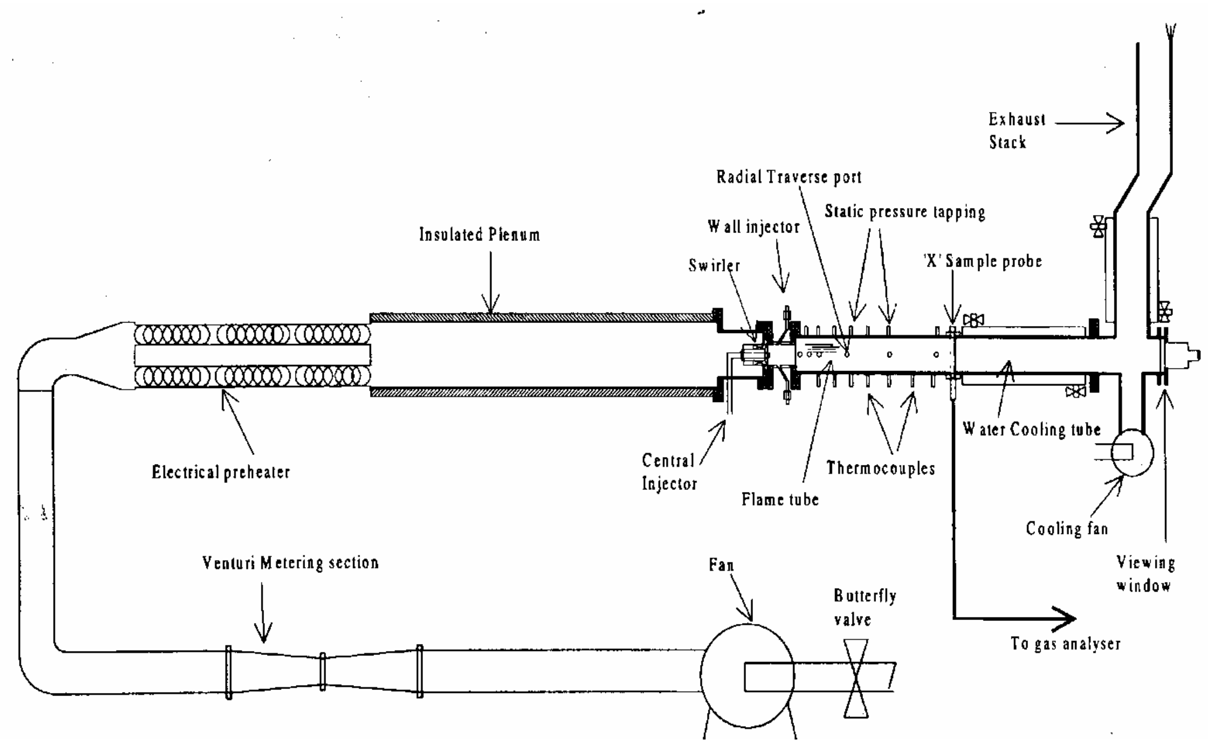

Figure1: Rig Test Facility

The inlet air temperature was measured 100mm upstream of the swirler using chromel-alumel type K thermocouple and the air flow rate was set at M=0.0225. This Mach number typically represents ~75% of the total combustor airflow entering the lean primary zone through the radial swirler. The ignition was carried out by electrical discharge from the spark igniters.

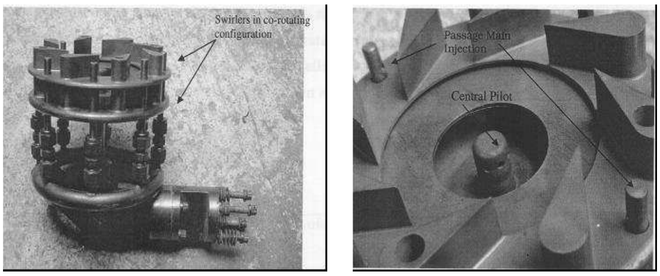

Figure 2: co-rotating radial double swirler with 25mm[9].

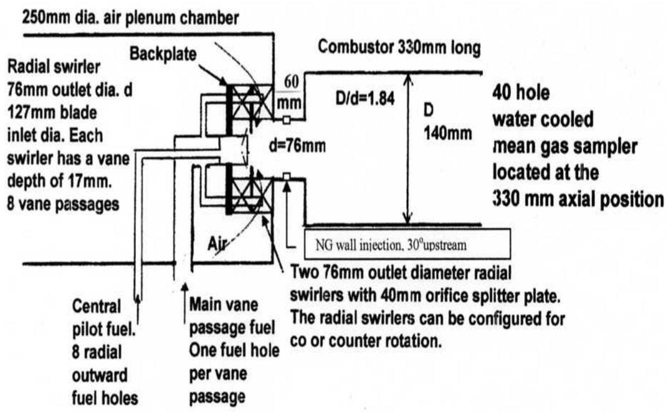

Figure 3: Combustor Geometry

Fuels and fuel delivery system

The WME was made from a mixture of waste cooking oils such as rapeseed oil and sunflower oil. Its physical and chemical properties are presented in Table1, along with kerosene. Kerosene was stored in a 200 liters barrel whereas WME and blends were stored in a 40 liter tank. They were pumped from the barrel or tank and delivered to fuel injection points after passing through rotameters for measuring fuel flow. Two rotameters were used with different measurement ranges. These two rotameters were calibrated for kerosene, WME and blends respectively as the density for these liquid fuels are different and thus the mass flow is different for the same indicated readings.

B100 (WME), B20 and Kerosene were injected and premixed with incoming air in a premixed fuel injector with holes on centers of equal area as shown in figure 2. The fuel air ratio and equivalence ratio was increased in small steps by increasing fuel flow rate while keeping air flow rate constant

Table.1 Fuel properties

Property	**Kerosene**	**WME**
Viscosity at 40°C mm^2/s	2.35	5.10
Density @15°C kg/m^3	800	884
Calorific Value MJ/kg	46	39.8
Stoichiometric ratio	14.7	11.8
Carbon wt%	85	75
Hydrogen wt%	15	11
Oxygen wt%	0	14

The experiment was started by supplying natural gas and igniting the methane-air reactant in the combustor. Then, the liquid fuel was gradually introduced to attain the desired value, while the methane flow rate was slowly decreased to zero.

Emission measurement

Regulated emissions

Mean exhaust gas samples were obtained using an 'X' configuration stainless steel water cooled probe with 40 holes at centers of equal area. The sample gases were passed into a 190ºC heated sample line and on through a 190ºC heated filter and pump to a 190ºC heated gas analysis system. The gas analyses results were processed to provide air fuel ratio. Regulated emissions (NO, NO_2, NOx, UHC& CO) have been analyzed on dry basis and corrected to 15% O_2 over a range of different fuel flow rates and reported separately

Unregulated emissions

A Gasmet CR-2000 Fourier Transform Infrared (FTIR) was used to detect and quantify three aldehydes: formaldehyde, acetaldehyde and acrolein, along with other 35 calibrated VOC compounds. The FTIR is capable of measuring concentrations as low as 3~5ppm with 2% accuracy of measurement range. The calibration ranges for formaldehyde, acetaldehyde and acrolein were 0~100, 0~200 and 0~500 ppm respectively. The exhaust sample was drawn at the same position as that for regulated emission analyzers by a heated sample line with its temperature being controlled at 180 °C. The sample flow rate was 2-3 l/min at a frequency of 0.5Hz. At least one hundred measurements for each testing condition were taken and the average values were employed. One of the main objectives of using the FTIR in this study is to get specified hydrocarbon information, including alkenes, aromatic hydrocarbon, and aldehydes [10]. This paper is focused on aldehydes emissions due to their strong ozone formation potentials.

Emissions Calculation

The gaseous emissions measurements were on a volumetric basis. These were converted into a mass basis using the conventional method for the computation of emissions index (EI: g/Kg fuel).

$$EI=K*C*(1+A/F)*1000 \qquad \text{(EI: g/Kg fuel)} \qquad 1$$

Where, K is conversion coefficient, which is the ratio of molecular weight of a certain emissions component to the molecular weight of the whole sample gas. The molecular weight of the exhaust sample gas is close to that of air and does not vary more than 1% for H/C ratios of about 2 irrespective to air/fuel ratio. For this reason, K is a constant per component.

C is the concentration of a pollutant in ppm

A/F is the air/fuel ratio.

Estimation of OFP (Ozone Formation Potential)

The ozone control strategy requires some quantitative methods to determine the tendency of ozone formation for a particular substance. The use of incremental reactivity is an appropriate reactivity ranking scheme. The incremental reactivity (IR) of an organic compound is defined as the amount of ozone produced per unit mass of VOC added to an organic mixture, or:

$$IR_i = \Delta O_3/\Delta VOC_i \qquad gO_3/gVOC_i \qquad 2$$

The maximum incremental reactivity (MIR) is the primary reactivity scale used in VOC control regulations for gasoline vehicles and aerosol coating in California [11, 12] and is developed using a box model. For the MIR box model calculation, NOx level is adjusted to result in the highest incremental reactivity. MIR conditions are likely to occur in VOC-limited urban air masses, where the mixture is most sensitive to organic compounds because of low VOC-to-NOx ratios. MIR represented the maximum possible ozone formation tendency under the optimum condition.

The EI of OFP for a particular VOC is calculated by the following equation:

$$EI\,(OFPi) = MIRi * EI_{VOCi} \quad gO_3/kgfuel \qquad 3$$

Where, EI_{VOCi} is the EI of VOCi in g/kgfuel.

The reactivity by definition is the sensitivity of ozone concentrations to the mass of individual organic compounds added to the system, and therefore, different sensitivity analysis techniques can be applied for their assessment.

The EU environmental law requires 31 individual OFP precursor VOCs and NMHC (Non-Methane Hydrocarbon) to be measured [13]. Formaldehyde is listed as recognition of its significance in OFP, in which MIR of formaldehyde is $8.96 gO_3/gVOC$. One of the objectives of the study is to assess the contribution of formaldehyde emissions to OFP.

RESULTS AND DISSCUTION

Weak extinction limit, stable combustion ranges and flame temperatures

Each fuel was tested to the lowest equivalent ratio (leanest) that could maintain a stable flame and to the highest equivalent ratio (richest) that acoustic noise became unbearable. For B100 case, the upper end of the curve (richest mixture) was constrained by the appearing of a large amount of white smoke from around the flanges of the combustor due to the evaporation of unburnt biodiesel. This indicated that the lower volatility of biodiesel, compared to kerosene, caused serious problems on fuel vaporisation and atomisation. Only three equivalent ratios were tested for B100 due to heavy smoke. The range of the equivalence ratio for fuel air mixtures of B100 was 0.3 to 0.5. The white smoke problems were removed by the addition of kerosene in the case of B20, The range of equivalence ratio of fuel air mixtures for B20 was increased as to 0.6~0.85. The 100% kerosene fuel showed the widest range of equivalence ratio with a value of 0.62 to 0.94. The weak extinction limit and rich fuel air mixture limit due to excessive smoke and acoustic level are listed in table 1 below. The B100 has a considerable lower weak extinction limit due to its oxygen content.

Table 1: Weak extinction limit and rich fuel air mixture limit constrained by smoke and acoustic noise

	Weak Extinction Limit phi	Maximum phi with bearable acoustic noise
B100	0.3	0.5
B20	0.6	0.85
Kerosene	0.62	0.94

The radial swirlers, as studied in the paper, are popularly used in industrial gas turbines. The design of swirlers and fuel injection location could affect emissions. Andrews et al investigated the four radial vane passage designs for low NOx lean combustion using natural gas at 600-740K and atmospheric pressure [14]. The four designs investigated were curved, flat bladed, rectangular and circular passage cross section. Each radial swirler had eight vane passages as in this work. Their results at various inlet temperatures and different fuel injection locations showed that the different radial swirler designs have small impact on weak extinction limit.

The flame temperatures were computed using GAS-eq software for three fuels as shown in figure 4 and showed a linear correlation with equivalence ratio irrelevant to fuel types.

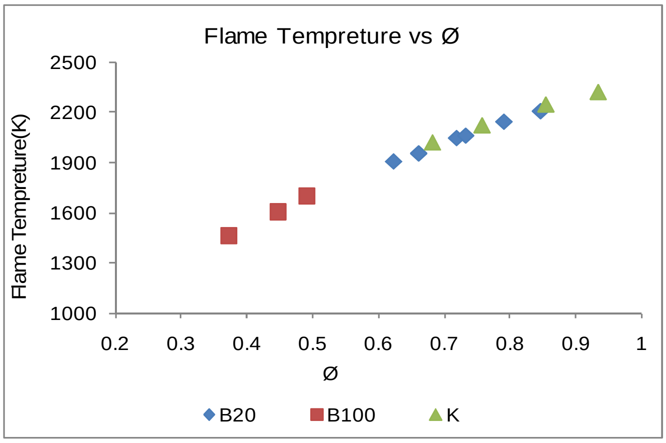

Figure 4: Calculated flame temperature Vs equivalence ratio for three fuels

Aldehydes emissions as a function of equivalence ratio

Only two FTIR measurements were taken for B100 at the equivalence ratio of 0.37 and 0.45. The effort for a FTIR measurement at the equivalence ratio of 0.49 was failed due to the heavy white smoke and consequently terminated the test.

The aldehydes measured by the FTIR are presented in ppm. Figures 5 to 7 represent three aldehyde species: formaldehyde (HCHO), acetaldehyde (CH_3CHO) and acrolein (C_2H_3CHO) in ppm as a function of equivalence ratio (Ø) for all three fuels under atmospheric pressure and 600K inlet temperature with Mach number 0.0225.

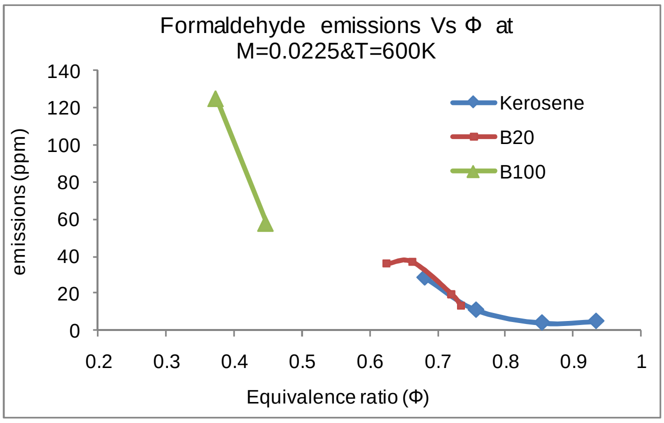

Figure 5: Formaldehyde as a function of equivalence ratio

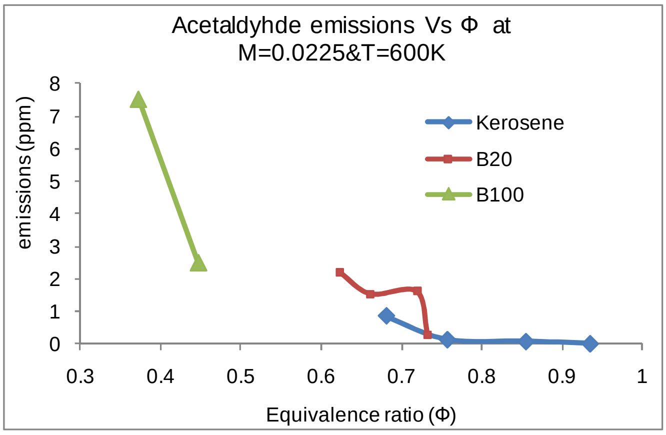

Figure 6: Acetaldehyde as a function of equivalence ratio

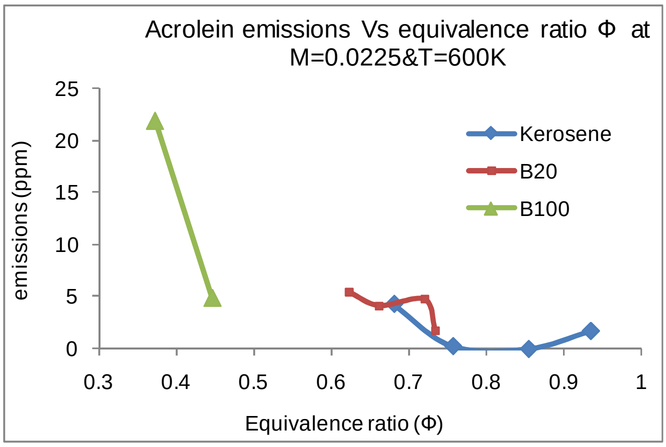

Figure 7: Acrolein as a function of equivalence ratio

The results showed that formaldehyde is the dominant aldehyde species for all three fuels. B100 had significantly higher formaldehyde emissions than B20 and kerosene. It is notably that the equivalence ratio for B100 was remarkably lower, which resulted in lower flame temperatures compared to B20 and kerosene (figure 4). This suggested that the flame temperature is a major factor that affects the formation of formaldehyde. The oxygen content in the fuel seems to have little impacts on formaldehyde formation by comparison of B20 with pure kerosene results where there was overlapped range of equivalence ratio. The same trends were found for acetaldehyde and acrolein. The concentrations of acetaldehyde were close to the detection limit of the instrument for B100 and lower than the detection limit for B20 and pure kerosene. There were notable acrolein emissions for B100 and very low levels for B20 and pure kerosene. The aldehydes are intermediate combustion products. Their formation is mainly due to incomplete combustion which is directly linked to combustion temperatures. The strong correlation between aldehydes and flame temperatures suggested that the formation of aldehydes were greatly decreased when flame temperatures increased to above 1600K.

The concentrations of aldehydes were converted into EI as a function of equivalence ratio for all three fuels as shown in figures 8 to 10. This will enable the estimation of mass emissions from fuel consumption.

The EI of formaldehyde for B100 from Ø=0.44 to Ø=0.37 is increased by a factor of 2.75 (1.6 g/kg fuel to 4.2g/kg fuel, larger than that of by concentration ppm (factor of 2.15, 58 ppm to 125ppm) as shown in figure 8. This is due to a larger exhaust flow rate at lower equivalence ratio than that at higher equivalence ratio. The similar differences appeared to acetaldehyde and acrolein. This emphasizes the importance of EI as a measure of emissions, especially at low equivalence ratio or high air fuel ratio.

The EI values of aldehydes for B20 and kerosene were much lower than that of B100. The peak value of formaldehyde emissions for B20 was (0.89g/kgfuel) at Ø=0.62 and decreases 0.3g/kgfuel as the fuel air mixture getting richer to reach Ø=0.73. The direct comparisons between different fuels were difficult due to different ranges of equivalence ratios. However, there was a small overlap range in equivalence ratio for B20 and kerosene at around Ø=0.7, where the B20 did not seem to have higher formaldehyde emissions.

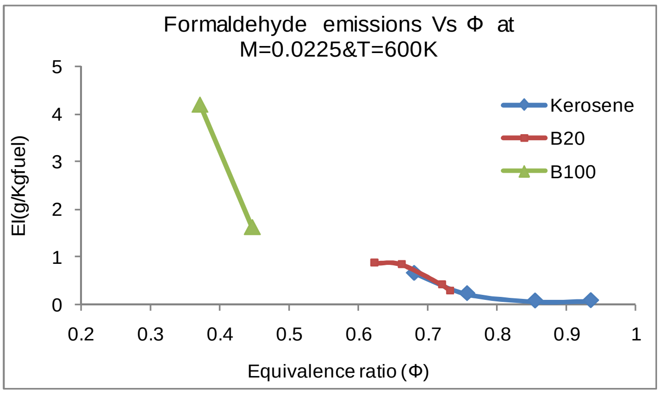

Figure 8: Formaldehyde as a function of equivalence ratio

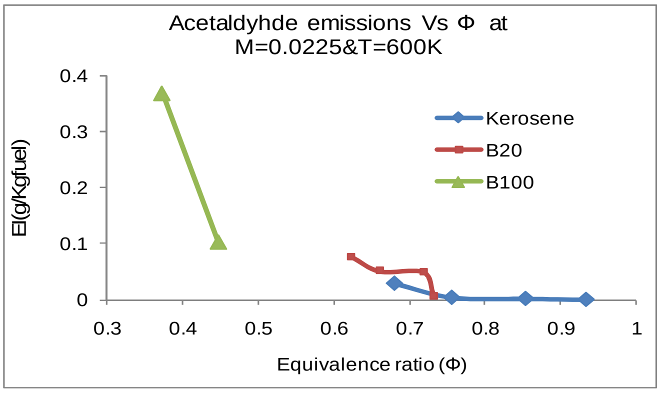

Figure 9: Acetaldehyde as a function of equivalence ratio

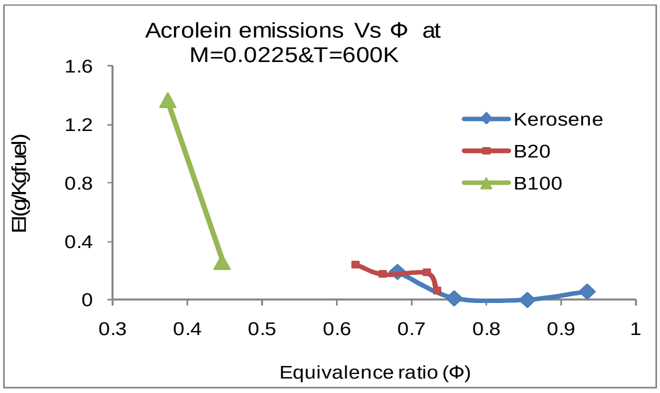

Figure 10: Acrolein as a function of equivalence ratio

<u>Formaldehyde emissions as a function of flame temperature</u>

Combining the resuts in figures 4 to 7, it is clearly shown that aldehydes emissions are a function of flame temperatures regardless of fuel types. It is therefore worth further investgating the relationship between the aldehydes concentrations Vs flame temperatures. As acetaldehyde and acrolein concentrations were very low at higher equivalence ratios, which contrained the accuracy of the measurement, only formaldehyde concentations were further invesigated. All formadehyde concentrations in ppm from three different fuels were plotted against flame temperatures as shown in figure 11. A good polynominal fit between formaldehyde concentration and flame temperature was shown. In fact, a good linear fit existed between 1600 and 2200 K. The same trend is shown in figure 12 for formaldehyde EI.

The formaldehyde emissions in ppm and EI were normalised to the maximum values as shown in figures 13 and 14. As the results represented a relative change from a particular equivalance ratio for all three fuels it could be used to predict other fuels such as B50 foramdehyde emissions trend based on limited measurements.

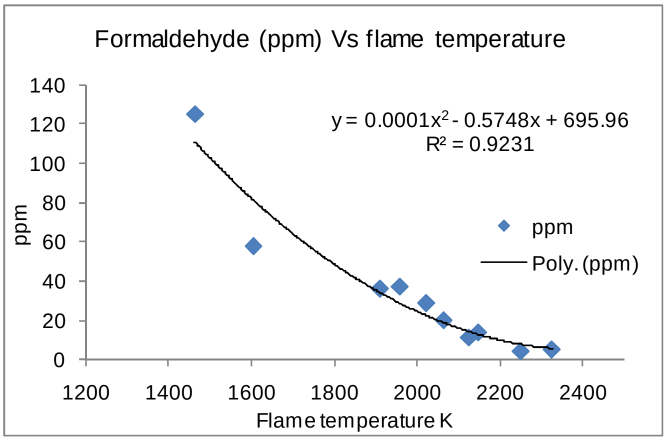

Figure 11: Formaldehyde concentrations as a function of flame temperature

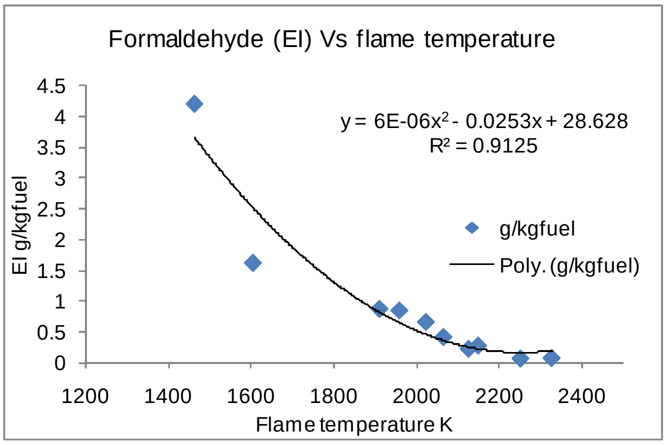

Figure 12: Formaldehyde EI as a function of flame temperature

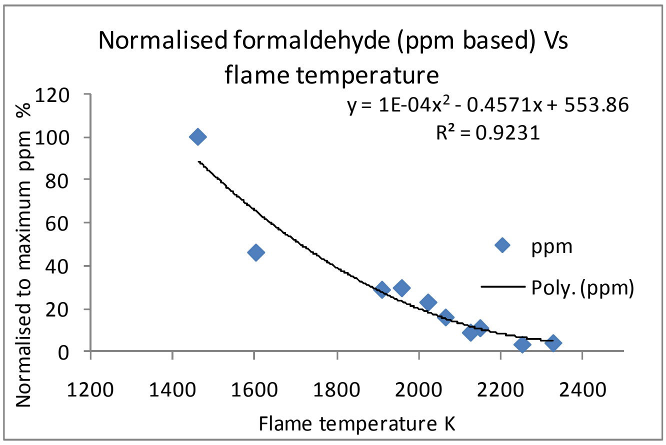

Figure 13: Normalized formaldehyde concentration as a function of flame temperature

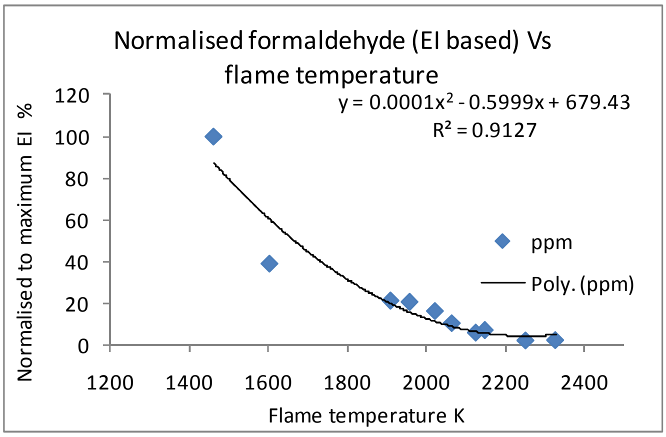

Figure 14: Normalized formaldehyde EI as a function of flame temperature

It is worth noting that industrial gas turbines for electrical power operate at high pressure around 20bar and aero gas turbines at take off could reach up to 60bar. The influence of pressure on emissions should be considered. The relation between the thermal NOx and pressure was reviewed by Andrews [15] and showed that the thermal NOx increased as the pressure rose. The strong influence of the pressure on NOx emissions occurred near stoichiometric region. The evidence for a ~0.5 pressure exponent for thermal NOx in lean burn combustion was observed. Liu etc investigated the biodiesel emissions on an industrial gas turbine and found that the CO and hydrocarbon emissions decreased as the inlet pressure increased [16], this may suggest that the increase of pressure leads to a reduction in aldehydes emissions. However, no literature or report on the influence of pressure on aldehydes emissions has been found. It is interesting to investigate the effect of pressure on aldehydes emissions in the future work.

Combustion characteristics of aldehydes

Salooja studied the role of aldehydes in combustion inside a furnace and investigated the pre-flame and ignition behaviors of a number of saturated and unsaturated aldehydes [17]. He concluded that:

1) Aldehydes vary in their influence on the oxidation reactions at different pre-flame stages and on ignition.
2) The aldehydes reactivity does not increase with molecular weight.
3) Acetaldehyde is the most effective promoter.
4) Formaldehyde powerfully inhibits combustion at all stages low and high temperature regions. This is due to a fact that formyl radical is stable and bond dissociation energy is much higher than that in the CH_3CO radicals.
5) The unsaturated aldehydes cause obvious inhibition in the early stage of reaction due to hydrogen atoms, whereas as in later stage the opposite scenario can be seen.

Wei et al [18] studied the aldehydes emissions mechanisms from blend fuel using spark ignition engine. They concluded that the emissions of formaldehyde and acetaldehyde are controlled by two factors: initial temperature and gas flow velocity (i.e. residence time). Also, no HCHO and CH_3CHO will be emitted when the temperature is too high or too low due to rapid consumption and the frozen generation respectively. In this study, the air flow rate was fixed and thus the residence time was constant. Therefore the aldehydes emissions were dependent on the consumption that would be accelerated at higher temperatures.

Total hydrocarbon emissions and aldehydes fractions

The total hydrocarbons (THC) for three fuels at different equivalence ratios were measured by a FID as presented in table 2, along with total aldehydes concentrations (converted to methane equivalent) and their fractions of total hydrocarbons. The THC emissions were similar for kerosene and B20 at all tested equivalence ratio ranges. The B100 had significantly higher THC emissions because of lower equivalence ratio and flame temperatures. Further increase of equivalence ratio would raise the flame temperature and then reduce hydrocarbon emissions for B100. However, the further increase of fuel flow for B100 was constrained by a large amount of white smoke due to inefficient fuel evaporation. This is very different from diesel engines where a much high fuel injection pressure is usually used and thus fuel spray and atomisation for B100 in diesel engines are in a much better situation. By comparison with B20 and kerosene, the results in table 2 shows that B20 produced more overall aldehydes emissions in terms of concentration and percentage of the THC at the similar equivalence ratio ranges, although the THC emission levels were similar for B20 and kerosene. The total ahdehydes emissions were considerably high for B100 and yet their percentages were not higher than other two fuels. For B20 and kerosene, the THC concentrations were at similar levels over the testing ranges of equivalence ratios, the concentrations and fractions of aldehydes were reduced significantly as the equivalence ratio increased. This indicated that as the fuel air mixture was getting richer, the flame temperature increased and aldehydes emissions were reduced while the THC remained. So the composition of the THC emissions changed.

Table 2: The percentage of Aldehydes to THC

	Ø	THC ppmC1	Aldehydes ppmC1	Aldehydes %
kerosene	0.68	100	43	43.2
	0.75	104	12	11.5
	0.85	109	4	3.7
	0.93	114	10	8.8
B20	0.62	104	56	54.1
	0.66	104	52	50.2
	0.72	104	37	35.7
	0.73	105	19	18.2
B100	0.37	680	206	30.3
	0.45	717	77	10.8

Assessment of OFP

The OFP of formaldehyde emissions at different equivalence ratios and flame temperatures were determined using equation 3 and results in figures 8 and 12 for three fuels and presented in figures 15 and 16. The results show the maximum possibility of ozone formation by formaldehyde emitted. The results in terms of gO_3/kgfuel enable the estimation of OFP as a function of fuel mass flow and thus total mass of OFP for a given fuel mass.

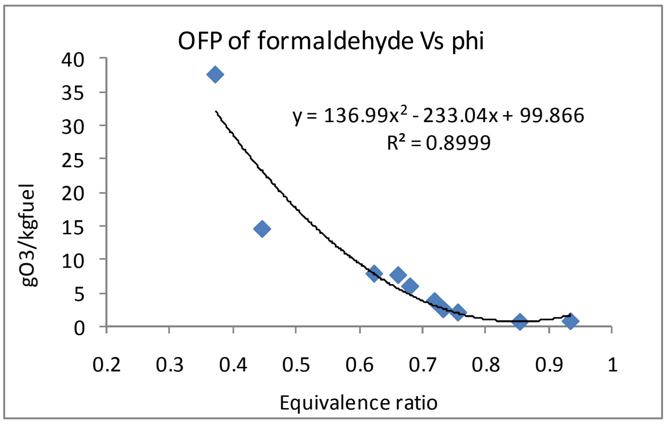

Figure 15: OFP of formaldehyde as a function of equivalence ratio for three fuels

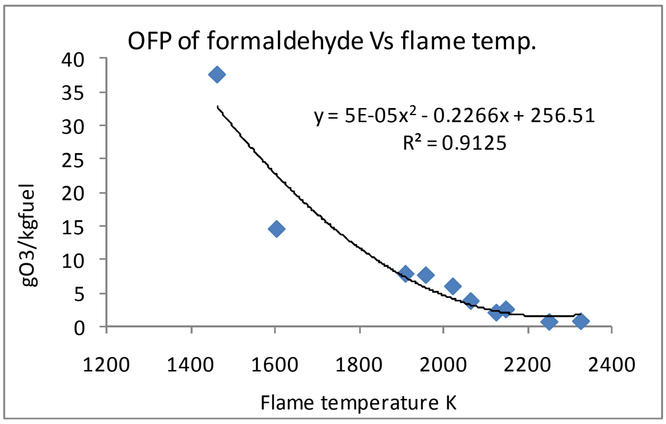

Figure 16: OFP of formaldehyde as a function of flame temperatures

CONCLUSIONS

The effects of pure WME (B100) and blend (B20) fuels on weak extinction limit, aldehydes emissions and OFP were investigated and compared with pure kerosene on a low NOx radial swirler combustor. The experiment results showed that the dominant aldehyde in the exhaust is formaldehyde, which could be accounted for up to 50% of the total hydrocarbon emissions for B20 at low equivalence ratios. Aldehydes emissions were reduced as equivalence ratio increased. This is attributed to increases in flame temperatures, which were found to have a significant influence on aldehydes emissions. There is a good correlation between aldehydes emissions and flame temperatures regardless fuel types, which could be potentially used for predicting aldehydes emissions for other fuel types based on equivalence ratios. The comparison between B20 and kerosene showed slightly higher aldehydes emissions in terms of concentrations for B20 than pure kerosene but with similar fractions of the total hydrocarbons. B100 produced much higher aldehydes, especially formaldehyde emissions at the low equivalence ratio, along with high total hydrocarbon emissions. The low volatility of B100 induced the fuel evaporation problem and confined the fuel air mixture within a very narrow equivalence ratio window and restrained the test towards higher equivalence ratios. The OFP of formaldehyde emissions was assessed and showed a strong correlation with equivalence ratio and flame temperature regardless of fuel types.

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