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The oxidative stability of synthetic fuels and fuel blends with monoaromatic blending components

Paul M. Rawson [a,c,*], Christy-Anne Stansfield [a], Renée L. Webster [a,b], David Evans [a], Ulas Yildirim [c,d]

[a] Defence Science and Technology Organisation, 506 Lorimer St, Fishermans Bend, Victoria 3207, Australia
[b] Australian Centre for Research on Separation Science, School of Chemistry, Monash University, Wellington Rd, Clayton, Victoria 3800, Australia
[c] School of Aerospace, Mechanical and Manufacturing Engineering, RMIT University, 124a Latrobe Street, Melbourne, Victoria, Australia
[d] Directorate General Technical Airworthiness, Royal Australian Air Force, Melbourne, Australia

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ABSTRACT

Alternative jet fuels and blends are required to meet a minimum aromatic concentration of 8%. It has been proposed that some monoaromatic compounds may make suitable single component aromatics to meet this minimum. One monoaromatic of interest is *p*-cymene which can be efficiently produced from eucalyptus oil. The oxidative stability of *p*-cymene and a range of monoaromatics blended with a range of alternative fuels are examined. Their oxygen induction times and peroxide formation under accelerated storage conditions are reported. It was found that some monoaromatics can impact final blend stability and that *p*-cymene produces very high levels of peroxides with all fuels examined. It was also observed that the choice of conventional fuel used for blending also influenced the blend stability.

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1. Introduction

With the increasing acceptance of alternative fuel blends by both commercial and military users there has been increased effort to understand the stability characteristics of synthetic fuels and blends made with fuels from conventional crude feedstocks. One mandated property of alternative fuels blends is a minimum aromatic content of the final blend of 8% by volume.

Aromatics are required in jet fuel to ensure compatibility with fuel seals to enable sufficient swelling characteristics [1]. Most conventional jet fuels used for blending are expected to have sufficient aromatic content to ensure a final conventional and synthetic blend will meet the 8% minimum. In situations where this is not the case and for potential future 100% synthetic fuels, an option for ensuring minimum aromatic content is the blending of aromatic compounds into the alternative fuel [2].

One proposed aromatic is *p*-cymene (1-Methyl-4-(1-methyle thyl)benzene) which can be produced by a pyrolytic conversion process using cineole extracted from eucalyptus oil as a feedstock. A process for this conversion is reported by Leita et al. [3]. This is an attractive option for indigenous Australian production due to the availability of eucalyptus feedstock and high yield of the conversion process.

As the current generation of alternative fuels are expected to be blended with conventional fuels up to 50% by volume, the low temperature (140 °C) oxidative stability of a range of blends with and without the addition of *p*-cymene and a range of alkyl monoaromatic compounds was examined. No diaromatics were chosen as they are currently limited to a maximum of 3% in jet fuel and are unlikely to be used in their pure form as aromatic blending components. Also the inclusion of diaromatics to synthetic fuels (synfuels) has been reported to negatively impact synfuel blend's thermal stability [4]. The synfuels available at the time of this study were experimental synfuels and not all met the requirements of ASTM D7566 standard specification for aviation turbine fuel containing synthesized hydrocarbons.

Fuel storage stability may be assessed by a range of techniques including induction times, peroxide formation and sediment formation [5–9]. Those chosen for this study were oxidation induction time and peroxide formation. The influence of aromatic blends on synfuel blend's thermal stability and sediment formation has been examined in detail by DeWitt et al. [4] using a range of aromatic distillate fractions as blending components and deposit formation as an indicator of stability.

It is expected that the influence of aromatic compounds on stability is based on their degree, relative position of alkyl substitutions and the presence of benzylic hydrogens. A range of

* Corresponding author at: Defence Science and Technology Organisation, 506 Lorimer St, Fishermans Bend, Victoria 3207, Australia.
E-mail address: Paul.Rawson@dsto.defence.gov.au (P.M. Rawson).

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monoaromatics were chosen to examine this influence and to specifically examine *p*-cymene's influence on stability.

Synfuels do not contain natural antioxidant capacity, which is normally derived from trace levels of compounds such as phenols and sulfur heteroatoms and their resistance to oxidative stress as measured by oxygen uptake rates is expected to be lower than conventional fuels [10,11]. It was expected that the naturally occurring antioxidant species in conventional fuels and the synthetic phenolic antioxidants added to hydroprocessed fuels may assist in stabilising the final blends. It has also been previously established that blending of stable and non-stable fuels may improve the final blend stability [12,13].

The distribution of individual aromatics is not normally characterised in jet fuel [14]. Aromatic content is reported only as a total percentage by volume or less often as a total aromatic broken down into mono, di and poly aromatic fractions.

This study examines the impact of individual mono-aromatic compounds on the stability of conventional and synfuel blends and the stability of synfuels blended with conventional fuels without further addition of an aromatic component.

2. Experimental

The following experiments were undertaken to examine synfuel blend stability and the influence of single monoaromatics on stability.

a. Blending of *p*-cymene at 0%, 8%, 16% and 24% v/v with a range of conventional and synthetic fuels with PetroOxy stability testing.
b. Blending of a range of individual mono-aromatics at 16% v/v with a range of conventional and synthetic fuels with PetroOxy stability testing.
c. Blending of conventional fuels with synthetic fuels without further addition of aromatics with subsequent PetroOxy and low pressure reactor stressing with peroxide testing.

To assist in understanding the impact of inclusion of high concentrations of a single component mono-aromatic, the natural abundance of the test aromatic compounds was measured for a set of jet fuels taken from in-service bulk tankage. All of the single component aromatics used in this study were found to occur naturally in the fuels examined.

2.1. Synthetic fuel samples

Synthetic fuels were received from a range of sources and are described in Table 1.

Samples of current generation alternative fuels were procured and where not available were generated. The butene oligomer and FT-GTL products were synthesized by local university research groups.

The FT-GTL synfuel was un-hydrotreated and contained approximately 75% *n*-paraffins, 5% iso-paraffins and 20% n-olefins of which 10% were 1-olefins. It was included to examine the impact of an un-hydroprocessed FT-GTL and what may be expected of its stability characteristics. All other synfuels samples were experimental fuels and not production batches. The synfuels synthetic phenolic antioxidant content was measured before blending. The results are reported using a gas chromatographic triple quadrupole mass spectrometry method [15]. A chromatogram of the synfuels is given in Fig. 1 to compare profiles which are each consistent with distillation characteristics of the respective fuels.

The two conventional jet fuels used for blending with the synthetic components were taken from known sources. The first from an in-service military base that was delivered from a refinery known to hydrotreat (JP-8-HT) and the second a Jet A-1 from a local refiner known to use a Merox finishing process (Jet A-1 – Merox).

Isopar M and dodecane were included in the trial to examine the response of the synthetic components to known *n*-paraffin and iso-paraffins that contained no antioxidant.

2.2. Monoaromatics

The single component aromatic compounds were obtained from Sigma–Aldrich (Castle Hill, NSW, Australia). Aromatics chosen where, *p*-cymene, *o*-xylene, ethylbenzene, 1,2,4-trimethylbenzene and 1,3,5-trimethylbenzene.

2.3. Experimental methods

Two methods were used to assess the fuel's storage stability. Induction time testing was undertaken by ASTM D7545 using a PetroOxy (Anton-Paar) instrument which determines a fuel's oxidation stability by heating 5 mL of fuel at 140 °C under an oxygen atmosphere. The stability result is the time taken for a 10% drop in the oxygen pressure. The PetroOxy is normally used for diesel fuels, however, when used for a range of jet fuels it has been found to have good repeatability for jet fuel induction period assessment at ±6%. This repeatability is based on our laboratory experience with repeated samples.

Blends were stressed and peroxide content measured based on a process described by Pande et al. using a low pressure reactor (LPR) at 100 °C with 500 kPa air overpressure for 16 h with subsequent peroxide measurement [16,17]. Peroxide testing was

Table 1
Summary of jet fuel samples used in this study.

Sample	Feedstock	Processing/refining method	Dominant component(s)	Total synthetic phenolic antioxidant, (mg/L)
Bioderived SPK	Camelina	Hydroprocessed esters of fatty acids	Mixed n/iso – paraffins from C8–C16	14.8
HT-IPK	Coal/syngas	High temperature Fischer–Tropsch	Mixed normal/iso paraffins C8–C16	12.3
HRJ	Algae	Hydroprocessed esters of fatty acids	Mixed normal/iso paraffins C9–C17	10.1
A2J	Biogenic isobutanol	Alcohol to jet	C12 and C16 iso-paraffins	NIL
FT-GTL	Natural gas	Un-hydrotreated Fischer–Tropsch	n-alkane + alkene mix C8–C17	NIL
Butene oligomer	Butene	Hydroprocessed oligomer	C12, C16, C20 iso-paraffins	NIL
Algae JP5	Algae	Hydroprocessed esters of fatty acids JP-5	C15–C18 *n*-paraffins plus iso-paraffins	NIL
DSH	Algae	Direct sugar to hydrocarbon	2,6,10-trimethyldodecane	NIL
Jet A-1 – Merox	Fossil	Merox	Conventional jet fuel	NIL
JP-8 – HT	Fossil	Hydroprocessed with military additives	Conventional jet fuel	14.2
Isopar M	Fossil	Purified synthetic iso-paraffins	Iso-paraffins	NIL
Dodecane	Fossil	Sigma–Aldrich	C12	NIL

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