RENEWELL

Drop-in biobrandstof via de katalytische hydrogenering van pyrolyse olie voor toepassing in de scheepvaart

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1. Ignition and combustion characteristics of hydrotreated pyrolysis oil in a combustion research unit, 2022.
2. Sustainable cyclo-alkanes from fast pyrolysis oil, IEA Task 34 – Pyne newsletter 48, July 2021

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3. Marine fuel from fast pyrolysis, IEA Task 34, Pyne Newsletter 52, december 2022
4. Engine operation on HPO-MGO blends, in prep.

Afkortingen/abbreviations

Abbreviation	Description
CRU	Combustion research unit
EGR	Exhaust Gas Recirculation
FPBO	Fast pyrolysis bio-oil
GHG	Greenhouse gas
HPO	Hydrotreated pyrolysis oil
HVO	Hydrotreated vegetable oil
LCA	Life Cycle Assessment
LCC	Life Cycle Costing
MGO	Marine gas oil
NO_x	Nitrogen oxides
PM	Particulate matters
RED	Renewable Energy Directive

Project gegevens

Projectnummer:	TBBE118001
Projecttitel:	Drop-in biobrandstof via de katalytische hydrogenering van pyrolyse olie voor toepassing in de scheepvaart
Penvoerder:	BTG Biomass Technology Group
Medeaanvragers:	Technische Universiteit Eindhoven & SeaNRG (GoodFuels)
Projectperiode:	01/01/2019 t/m 30/04/2022
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Publieke Samenvatting

De Europese Commissie heeft de ambitie uitgesproken om de transportsector in zijn geheel te verduurzamen door bijvoorbeeld elektrificatie en gebruik te maken van hernieuwbare brandstoffen. In het geval van de luchtvaart en scheepvaartsector kan een vloeibare duurzame drop-in brandstof de voorkeur hebben, aangezien er hierdoor geen veranderingen nodig zijn in bijv. infrastructuur, distributie en eindgebruik. Momenteel zijn commercieel verkrijgbare duurzame brandstoffen voornamelijk gebaseerd op plantaardige oliën & vetten, en een uitbreiding naar RED2-conforme tweede generatie (2G) biobrandstoffen is hier gewenst.

Het specifieke doel van het Renewell-project was het ontwikkelen van een proces voor de productie van een duurzame, 2G drop-in scheepsbrandstof. Deze moet dan geschikt zijn om ten minste 5% van de conventionele scheepsbrandstof (MGO) te kunnen vervangen, en met de ambitie om 30% vervanging te bereiken. De "Renewell-route" is een meertraps-proces gebaseerd op snelle pyrolyse. Eerst wordt de biomassa omgezet in een mineraalvrije pyrolyse bio-olie (FPBO). Vervolgens wordt de FPBO bij verhoogde temperatuur en druk met waterstof behandeld gebruikmakende van verschillende katalysatoren om een Hydrotreated Pyrolyse-Olie (HPO) te verkrijgen. De chemisch-fysische eigenschappen en verbrandingseigenschappen van zowel de HPO als de HPO/MGO-mengsels worden dan in detail geëvalueerd. Renewell is een samenwerking tussen BTG Biomass Technology Group BV, de Universiteit van Eindhoven en Goodfuels Marine BV.

De drop-in biobrandstoffen zijn geproduceerd door BTG op lab- en pilotschaal. De hout-gebaseerde pyrolyse-oliën zijn verkregen van commerciële FPBO-productie-installaties (Empyro, GFN). Daarnaast zijn specifieke pyrolyse-oliemonsters (o.a. miscanthus, pyrolytische lignine, pyrolytische suikers) in het project gebruikt die door BTG geproduceerd zijn. De FPBO werd gestabiliseerd gebruikmakend van een gepatenteerde katalysator (Picula™) en verder opgewaardeerd met behulp van commerciële verkrijgbare katalysatoren. De verkregen HPO's zijn zowel intern als door derden in detail geanalyseerd. Over het algemeen kan worden geconcludeerd dat de HPO's aan alle specificaties van (destillaat) scheepsbrandstof voldoen met uitzondering van het vlampunt. Een te laag vlampunt werd veroorzaakt door de aanwezigheid van lichte componenten (met name cyclohexanen). Door het verwijderen van deze lichte componenten kon het vlampunt worden verhoogd tot 60 °C, dat het minimum is voor scheepsbrandstoffen.

Het verbrandingsonderzoek is uitgevoerd aan de Universiteit van Eindhoven. In eerste instantie zijn de HPO's en mengsels onderzocht/getest in een zogenaamde Combustion Research Unit (CRU). De HPO/MGO-mengsels vertoonden vergelijkbaar eigenschappen als diesel met hierbij een toenemende ontstekingsvertraging bij toenemend HPO-bijmeng percentage. Een mengsel van 75% HPO en 25% HVO (Hydrotreated Vegetable Oil) – een 100% hernieuwbare brandstof – vertoonde dezelfde verbrandingskarakteristieken als EN590-diesel. Blijkbaar wordt het relatief lage cetaangetal van de HPO gecompenseerd door het hoge cetaangetal van HVO.

De motortesten zijn uitgevoerd op een HD-dieselmotor testopstelling, een aangepaste DAF 6-cilinder MX13 motor. Tijdens de motortesten werd alleen de eerste cilinder gebruikt als de testcilinder, terwijl de overige vijf waren uitgeschakeld. Bij gebruik zonder EGR (=rookgascirculatie) zorgden zowel diesel- als HPO-brandstoffen voor ultra lage PM-emissies (fijnstof) van de motor. Toch namen de PM-emissies

toe terwijl de NOx-emissie afnam naarmate de HPO-mengverhouding toenam. Een goede beheersbaarheid en respons van het verbrandingsproces werd waargenomen wanneer het injectiemoment/timing en de brandstofdruk werden gevarieerd. Deze relatie tussen PM/NOx-emissie werd zowel waargenomen voor diesel als voor de HPO/MGO-mengsels. De verwachting is dat de NOx-emissie aanzienlijk kan worden gereduceerd wanneer de EGR wordt toegepast.

Het Renewell-project heeft aangetoond dat het mogelijk is om de motor te laten draaien op een 2G drop-in biobrandstof geproduceerd uit FPBO. Het is gebleken dat de motor veilig en soepel kan lopen op een mengsel van ten minste 30% HPO/MGO zonder grote invloed te hebben op de verbrandings- en emissiekarakteristieken. Op basis van deze positieve resultaten zijn er -na afloop van het project-testen op 50% HPO/MPO brandstofmengsels, en ook deze bleken probleemloos te verlopen.

Uit de uitgevoerde duurzaamheidsanalyse bleek verder dat, in vergelijking tot fossiele scheepsbrandstoffen, een behoorlijke broeikasgasemissiereductie kan worden behaald met de HPO geproduceerd uit FPBO. In een screening LCA bleek dit 85% tot 89% te zijn en 63% gebruikmakende van de RED II-methodiek. De uitstoot zou nog verder kunnen worden gereduceerd als een duurzame waterstofbron zou worden gebruikt in de productie van de HPO. Een economische beoordeling/assessment toonde verder aan dat de HPO geproduceerd uit FPBO kan concurreren met andere 2G-biobrandstoffen.

Sustainable cyclo-alkanes from fast pyrolysis oil

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Jet fuels consist of n-alkanes, iso-alkanes, cyclo-alkanes and aromatics. From a com-bustion point of view the aromatics are not desired, but its presence might be required in view of its seal swelling capacity. Cycloalkanes may have the same functional benefits as the aromatics, but they have a higher specific energy and likely their combustion results in lower particle emissions [1].

Jet fuels like JP9 and JP10 are speciality fuels for very demanding applications, and for example JP-9 contains methyl-cyclohexane to achieve the required specifications. Methyl-cyclohexane and sometimes ethyl-cyclo-hexane are included as a model component in jet fuel surrogate blends used in research activities. Therefore, cyclo-alkanes produced from renewable resources might be an interesting additive to jet-fuel to improve its sustainability.

Hydrotreatment of fast pyrolysis bio-oil (FPBO) can be a promising approach to obtain sustainable cyclo-alkanes from renewable resources. FPBO can be produced from a variety of biomasses and residues, and the technology is maturing and nowadays implemented commercially [2]. However, the FPBO as such is not suitable for direct use as transportation fuel; for example, it is acidic, contains significant amounts of water, and has a relative low heating value compared to fossil fuels. Obviously, severe upgrading will be needed, and typically, a two-step hydro-treatment process is applied to change the functionalities and remove the oxygen. The hydrotreatment process is carried out at elevated pressure and temperature in the presence of a catalyst.

In the Dutch funded project called *Renewell* the upgrading of FPBO is further developed with the objective to produce a drop-in, distillate marine fuel. Compared to aviation fuel the specifications of marine fuel are less strict and initial implementation is expected to be easier.

In the Renewell project FPBO is first stabilized over the proprietary Picula™ catalyst at a pressure of 200 bar and temperatures of $100 - 300\,°C$. The product from this first step is called Stabilized Pyrolysis Oil (SPO). In the second step the SPO is further treated over a conventional, sulphided NiMo catalyst at temperatures up to $450\,°C$ and pressures in the range of $100 - 120$ bar. Depending on the severity of treatment the products are called Stabilized Deoxygenated Pyrolysis Oil (SDPO) or Mixed Transportation Fuel (MTF).

Figure 1: Process Development Unit for the pyrolysis oil hydrotreating

Table 1: Flashpoints of the fuel before and after distilling off the lights

Fuel	Flashpoint [°C]		Lights removed [wt %]
	Before	After	
SDPO (wood oil)	< 12	64	14
MTF (wood Oil)	< 12	64	26
MTF (pyrolytic lignin)	< 12	81	52
MTF (Pyrolytic sugar)	< 11	61	35

In most cases wood based FPBO is used as feedstock, but for example also FPBO from miscanthus or specific fractions of FPBO (extractives, lignin or sugars) are tested. For the experimental work four labscale hydro-treaters are available as well as a process development unit (PDU). The latter one has a capacity of 20 – 50 kg input per day.

The chemical/physical properties of the fuels produced have been compared to marine distillate fuels. Specifications are met with respect to e.g. heating value, acidity, sulphur content, density and viscosity. Generally, the fuels blend very well with conventional fossil fuels and no phase separation is observed. Research on the combustion properties is on-going at the University of Eindhoven, and first results are very positive. Currently, the critical property to qualify as a true drop-in marine fuel is the flashpoint. This flashpoint is an important property for transport, safety, and storage; for marine fuel the minimum value is 55 – 60 °C.

The MTF contains light flammable components resulting in a low flashpoint and values around 10 °C have been measured. The light components should be removed from the MTF in order to increase the flashpoint to an acceptable level. This has been achieved by a simple distillation of the MTF removing the lights and evaluate the effect on the flashpoint (trial & error approach). In Table 1 the results are shown for some samples.

After distilling off the lights the fuel properties comply with the specifications for a marine distillate fuel. The question is then what is the composition of the light fraction and how can this fraction be used.
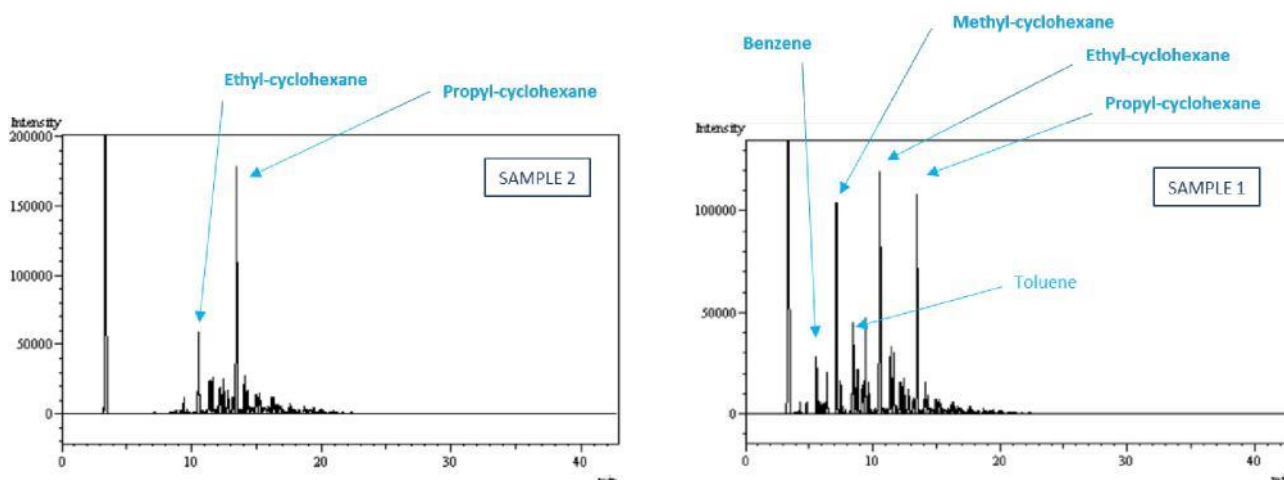

Fig. 2: Light distillates: chromatograms of GC-FID analysis of sample 1 and 2

The light fraction is a transparent liquid which burns very easily, and the flashpoint of this fraction is below zero. To get more insights in the composition of these light distillates a more detailed analysis by GC-FID was performed.

In the example below distillate fractions obtained from wood based MTF were further analysed. The first sample (sample 1) is obtained at a temperature of 85 – 100 °C, the 2nd sample (sample 2) is the fraction obtained by further increasing the temperature to 120 °C. In both cases the pressure was 100 mbar. Subsequently, the samples were dissolved in IPA and analysed by GC-FID.

The analysis of sample 1 is illustrated in the chromatogram in figure 1 at top right; the chromatogram on the bottom right corresponds to sample 2. By comparing previous GC-analysis of other samples and comparing retention times, a prediction could be made of main components present in the distillates.

Mainly methyl-, ethyl- and propyl-cyclohexane's were identified in the distillate obtained at 85 – 100 °C, while propyl-cyclohexane was mainly found in the distillate obtained at 100 – 120 °C. This was confirmed by the University of Groningen who analysed the samples by GC-MS. The cyclo-hexanes found in the distillates have ascending flashpoints with increasing (substituted) alkyl length, starting from -4 °C to 35 °C. Besides the cyclo-alkanes, benzene, toluene and octane were detected in the sample. Absolute amounts of the cyclo-hexanes in the samples were around 15 % Methyl-cyclohexane, 30 % Ethyl-cyclohexane and 35 % Propyl-cyclohexane. However, the current focus is on the production of a drop-in marine fuel, and the process nor the feedstock is optimised to produce cyclo-hexanes. For example, the percentage of lights seems to be much higher when miscanthus derived FPBO is used instead of wood derived oil meaning that it might be a better source for cyclo-alkanes. Furthermore, the crude mix of cyclo-alkanes produced here could be further separated and purified and used as a sustainable additive to jet fuel.

References

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Acknowledgement

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Full Length Article

Ignition and combustion characteristics of hydrotreated pyrolysis oil in a combustion research unit

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ABSTRACT

Biomass-derived fuels are promising in reducing life-cycle CO_2 emissions and achieving the goal of sustainable mobility in the future. This work investigates the ignition behavior and combustion process of hydrotreated pyrolysis oil (HPO) derived from various biomass resources. They are tested in a combustion research unit based on constant volume combustion technology, which imitates the ignition behavior in compression ignition en-gines. Various conditions are tested and HPO are benchmarked with commericially avalable biofuels and fossil fuels: hydrotreated vegetable oil (HVO) and fatty acid methyl ester (FAME), diesel, and marine gas oil. The results showed that the igntiion delay time follows an order of folloing: HPO > diesel-like fuels > HVO. Both the biomass type and after-treatment have a small influence on the ignition delay of HPO. Two combustion regimes are observed at different chamber temperature range. It also revealed that blending HPO into HVO can extend the ignition delay of HVO. And ignition delay of HVO/HPO increases as the HPO blend ratio increases. At 75 vol % HPO blend ratio, the HPO/HVO blend shows identical ignition and combustion behavior as diesel. In addition, the viscosity of HPO/HVO blends and diesel are also quite similar. The results indicated the possibility of using 100% bio-fuel in a modern marine engine to provide power to future mobility.

1. Introduction

Bioenergy production is expected to increase from 9.7×10^6 to 4.6×10^7 GJ d^{-1} between 2016 and 2040 [1] and biofuel is definitely a crucial part of it. In the transportation sector, measures have to be deployed to increase the efficiency of the transport system and cooperation should be reached globally to speed up and scale up the application of low-carbon alternative energy and moving towards carbon–neutral vehicles. The European Commission (EU) proposed the minimum 2030 target for the share of renewable energy consumed in the union should be 32%. Fuel suppliers are also required to ensure 14% of transport fuel originates from renewable sources [2]. Specifically, the implementation of biofuels in heavy-duty (HD) and marine transportation is one of the technical routes to cope with current CO_2 policies.

The production and application of biofuels will determine their overall environmental impacts. After decades of research and develop-ment, biofuels have experienced iteration of three generations. First-generation (G1) biofuels are mainly from edible agricultural crops such as corn, sugar cane, and vegetable oil [3]. Typical G1 biofuels are ethanol, fatty acid methyl ester (FAME), and pure plant oil (PPO). FAME is generally termed biodiesel, made via transesterification. With similar biomass sources, HVO is processed with hydrotreatment, which elimi-nates the oxygen content in the feedstock and saturates the long carbon chain compound [4]. These treatments also decrease the double/triple

carbon bond and make paraffins the main component of HVO. In addi-tion, HVO is found to show near-zero aromatics and shorter ignition delay than diesel. Second-generation biofuels are produced from none-edible biomass such as energy crops, cellulosic waste, agriculture, and forest residues. The raw materials include miscanthus and short rotation coppice (SRC) willow, wheat straw, and woody biomass [5]. Therefore, they are also called biomass-to-liquid (BTL). Examples for G2 are methanol, di-methyl ether (DME), 2,5-Dimethylfuran (DMF), and butanol [6]. Though both GI and G2 biofuels have received potential awareness for conflicting with agricultural land usage, G1 biofuels are more in the spotlight and raise more ethical issues in the 'food vs fuel' discussion. Despite their reputation, the current biofuel market is dominated by the first generation as a result of their mature production process. Almost 50 billion liters of biofuels have been produced annually by Shell since 2010 [7]. The cost depends on the biomass type, process strategy, production scale, etc. For example, the HVO plants are commercially available at 0.05 to 1 million (metric) tons per year output. The estimated cost of production falls in the range of 600−1100 EUR/ton (or approx. 50−90 EUR/MWh) [8]. While production cost of ethanol from lignocellulosic sugar via fermentation is about 103 EUR/ MWh. Compared to the G1 biofuels, the production of the G2 biofuel is more complex and remains at a developmental stage and not available on a commercial scale [9]. It requires a large investment in technology for mass production. The recent construction cost of a cellulosic ethanol plant with an annual capacity of 113 million liters, totaled $225 million. In contrast, the investment required for a corn ethanol plant that pro-duces 150 million liters per year is about $80 million [10].

Extensive work has been done regarding the combustion and emis-sion characteristics of these G1 and G2 biofuels, including the mea-surements in internal combustion engines, constant volume combustion chamber (CVCC), optical setups, and stationanry burners facilities. Li et al. [4] investigated the combustion characteristics of HVO, tallow methyl ester (TME), and ultra-low sulfur diesel (ULSD) at various blends in a CVCC facility. It was reported that HVO showed the shortest ignition delay and longest burn duration. The authors also found that the increased addition of ULSD in other fuels shortened the ID. Moreover, it was observed that the transition from diffusion-dominated combustion to premixed-dominant combustion as the chamber temperature de-creases. Van de Beld et al. [11] shaded lights on the potential of using the fast pyrolysis bio-oil (FPBO) in diesel engines for combined heat pow-ertrain (CHP). The tests were carried out in a single-cylinder, 20 kW, diesel engine. The results showed that the FPBO can be successfully combusted applying 100−120 ℃ inlet temperature with an engine compression ratio (CR) of 17.6. This inlet temperature can be further decreased by 40 ℃ as the CR increases to 22.4. As a result of the low energy density and high water content of FPBO, relatively low peak temperatures were observed which is corroborated by the fact that CO emissions were higher but NO_x emissions were reduced. It was further concluded that no notable effects on flue gas emissions and fuel con-sumption were noticed after 40 hours of durability tests. However, FPBO can not be considered as a drop-in fuel as it is not compatible with fossil fuels like diesel or heavy fuel oil (HFO).

Rogalinski et al. [12] compared the combustion properties of light fuel oil (LFO), HFO, rapeseed oil (RSO), and Glycerol under various thermal dynamic conditions in a combustion research unit. The results revealed that HFO presented the longest ID due to the lower cetane number (30) compared to that of RSO (41) and LFO (50). Specifically, the ID of RSO was both significantly influenced by chamber pressure and temperature while ID of HFO was most affected by chamber pressure. With pilot injection applied to enrich the air/fuel mixture, the ID of glycerol was relatively short. It was also reported that injection pressure had more effects on ID than injection duration because of its direct in-fluence on atomization. The authors also concluded that the ID at low ambient temperature and conditions was also influenced by the kinetic viscosity of fuels. Galle et al. investigated the physical properties of rapeseed methyl esters (RME), animal fat (AF), two straight vegetable oil

(SVO), and rapeseed oil (RSO) under non-evaporating conditions in an optical combustion chamber [13]. High-speed cameras were adopted to study the spray penetration and spray angle. Although haivng widely differnet biomass sources, they all presented higher viscosity, density, and bulk modulus than regular diesel. Among these biofuels, significant differences of the aforementioned physical properties were reported. The authors also indicated that the composition differences among these biofuels have a larger influence on the ignition and combustion than the atomization. The results have shown that the fuel temperature was an important parameter to control because it significantly affects the fuel properties. They have a large effect on both the injection timing and injection duration. While spray development was much less influenced by these properties. At low temperatures, a strongly deteriorated at-omization of oils and fats was observed. Xu et al. studied the laminar burning characteristics of a fast pyrolysis biofuel in a CVCC facility [14]. Catalytically produced from rice husk, 96% of this biofuel is made up of ethanol, ethyl acetate, diethyl ether, acetone, and 2-butanone with a mass ratio of 9:6:2:1:1. To investigate the spherical propagating flame, tests were conducted at initial pressures of 0.1−0.4 MPa, initial tem-peratures of 358−418 K, and equivalence ratios of 0.7−1.4.S_u increased at the early stage of combustion when both temperature and pressure increased. While S_u peaked at around $\varphi = 1.0$−1.1 and then decreased up to the point where cellular flames appear. The work carried out by Xuan et al. fills the research gap of combustion characteristics of compressed-ignition sprays with a gasoline-hydrogenated catalytic biodiesel blend [15]. 40% hydrogenated catalytic biodiesel was blended with gasoline and it was concluded that the liquid length, ignition delay, and lift-off length are enlarged for the cooling mode compared to that of the un-cooled one. The cooling jacket of the injector also brings in a larger overlapping area between liquid length and flame lift-off length. How-ever, the in-flame soot production for the cooling jacket is increased more than twice compared to that of the uncooled one.

The so-called third-generation (G3) biofuels are generally produced from micro-organisms, which are photosynthetic such as microalgae and microbes. Compared to conventional terrestrial feedstocks (G1 and G2 biofuels), they inherently pose advantages, such as higher growth ten-dencies, constant yield throughout the year, tolerance to high CO_2 content and harsh habitat conditions, and 30% higher lipid composi-tions than soybeans and palm oils [16]. Typical G3 biofuels include methane, bio-ethanol, bio-butanol, biodiesel, and bio-hydrogen [17]. Due to the complexity and high cost, G3 biofuels production is currently on small scales only and are not commercialized yet. But there have been some sources reporting the price of microalgae biodiesel in the range of $0.63/L (best case scenario with highly optimistic lipid yields) [18] to $2.60/L [19]. Quite a few studies exist regarding the usage of G3 biofuel in internal combustion eigne with a particulat interst in advanced combustion concepts, such as partially premiexed combustion (PPC) [20,21] and reactivity controlled compression ignition (RCCI) [22,23]. For example, the experimental work conducted by Seykens et al. [24] revealed the application of short-chain alcohol derived from seaweed in RCCI mode on HD diesel engines. Specifically, E85 and neat n-butanol were used as low reactivity fuel (LRF) via port-injection respectively and regular diesel was used as the high reactivity fuel (HRF) through direct injection. Tests were conducted both on single-cylinder HD engine setup and 6-cylinder HD engine setup. The results show that E85/diesel RCCI achieved 52% indicated thermal efficiency (ITE) in single-cylinder en-gine measurements and 46.6% ITE in 6-cylinder engine measurements. While the gross ITE for n-butanol/n-heptane RCCI was 51.6% found on the single-cylinder tests. The presented results demonstrated the po-tential of seaweed-based fuels is an important driver for upscaling the production process of these fuels. Some papers compared the production and application of FAME and hydrothermal liquefaction (HTL) biocrude from microalgae. Specifically, the FAME is mainly produced from dry mass, i.e. extracting the lipid (using solvent) and transesterification [25]. While HTL converts the whole biomass into crude oil [26]. Microalgae FAME presents similar properties as diesel and is suitable for
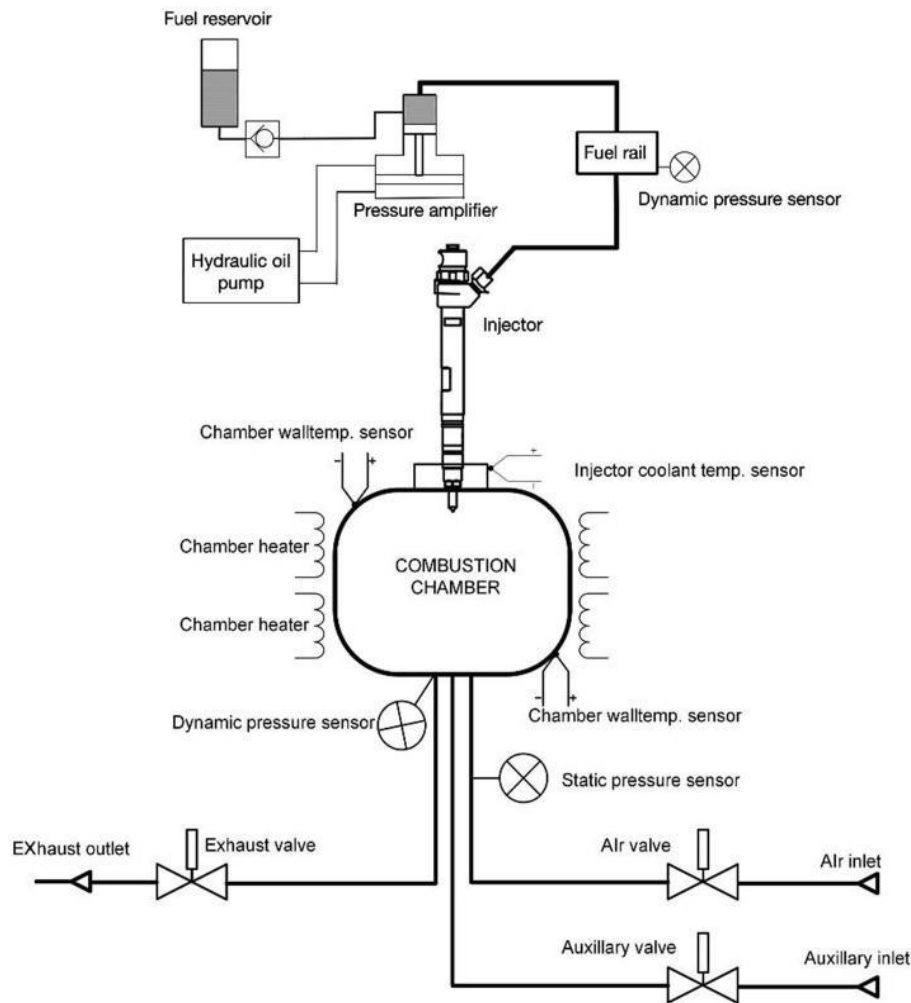
Fig. 1. Schematics of combustion research unit, reproduced with the permisison from Fueltech [32].

high-speed diesel engines including buses, tractors, cars, and similar vehicles. Whereas HTL biocrude has properties like heavy fuel oil (HVO) and is suitable for low-speed diesel engines, such as large marine ship engines and large electric generators [27]. Research work by Karthi-keyan et al. showed the overall performance of a CI engine fuelled with biodiesel/diesel blends [28,29]. The biodiesel is produced from the S. Marginatum macroalgae. It was reported that physical and chemical features of biodiesel/diesel blends (0%, 20%, 50%, 75%, 100%) were identical to regular diesel according to ASTM D6751. Performance and emission tests were performed on a single-cylinder diesel engine (compression ratio: 17.5, displacement: 0.66 L) at 1500 RPM. The au-thors reported that 20% biodiesel/diesel blends (B20) present the highest thermal efficiency, the lowest brake fuel consumption, and the highest exhaust temperature. Though the NO_x emissions for B20 in-creases, B20 yielded the lowest smoke opacity and CO/HC emissions. Rajak et al. [30] compared the performance and emissions of regular diesel and algae-biodiesel in a common-rail direct injection diesel en-gine at various CR (16.5–18.5). The numerical results from Diesel RK model showed that 17.5 was found to be the most optimal CR. In addition, it was reported that fuelling the engine with algae-biodiesel decreased brake thermal efficiency by 2.73%, torque by 6.66%, exhaust gas temperature by 1.6%, CO_2 by 6.1%, NO_x by 0.5% and par-ticulate matter by 60%. And the specific fuel consumption at CR 17.5 was increased by 6.4% as compared to diesel. Chen et al. [31], compared the combustion and emission characteristics of a dual-fuel engine fueled with diesel/methanol and diesel-polyoxymethylene dimethyl ether (PODE) blend/methanol at 4.2 bar, 7 bar, 9 bar BMEP. Particularly,

methanol was introduced by port-injection with an output substation ratio (MSR) of 20% and 40%. 50 vol% PODE was pre-blended with diesel (P50) and directly injected into the cylinder to investigate the effect of PODE addition. It was reported that P50/methanol showed a higher peak cylinder pressure, lower first peak heat release rate (HRR), and higher second peak HRR than the dual-fuel engine fueled with diesel/methanol. Both the ignition delay and combustion duration of the dual-fuel engine decreased when the pilot fuel injection changed from diesel to P50. Moreover, both NOx emissions and particulate matter produced by the P50/methanol engine were lower than those from the diesel/methanol engine for the specific MSR and engine load.

The above-mentioned literature shows the research work for different generations of biofuels has been prevailing in both academic and industrial research fields owing to the consideration of low life-cycle CO_2 emissions. It can be stated that biofuels will play a crucial role in future mobility, together with other energy carriers (i.e. batteries and none-carbon fuels). Though some interesting work has been done regarding the characteristics and application of specific biofuels. More comprehensive and dedicated experimental investigations are necessary before new biofuels are introduced into the market either as component or alternative fuel. This paper focuses on the ignition and combustion process of a hydrotreated fast pyrolysis bio-oil (HPO). Experimental work is done on a combustion research unit (CRU) based on CVCC technology. The combustion property of HPO derived from different biomass sources are benchmarked against commercially available bio-fuels and fossil fuels: FAME, HVO, diesel (EN590), marine gas oil (MGO) at various ambient conditions. With an ambition to use HPO as drop-in

Table 1

CRU basic specifications.

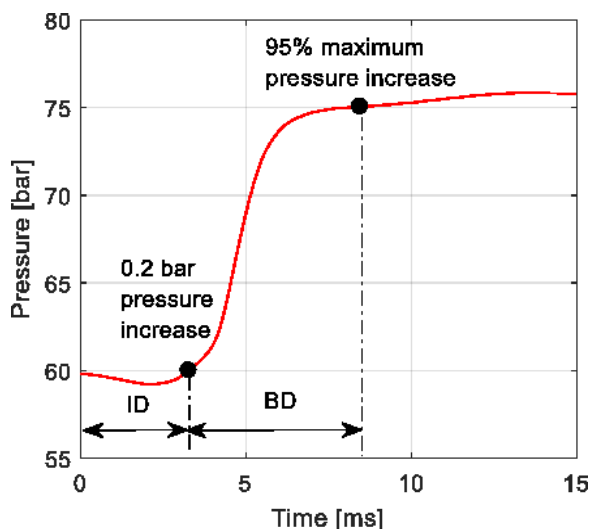| Parameters	Range	Limits
Chamber volume	475 [cm³]	
Chamber temperature	300–580 [°C]	Ref ± 1 °C for 150 s
Chamber pressure	10–60 [bar]	Ref ± 3.0 bar
Injection duration	0–1.5 [ms]	

Fig. 2. Chamber pressure as a function of time. A graphical interpretation of ID and BD is included.

fuels in marine applications without major modification, HPO was also blended with HVO in various blend ratios. Ideally, the physical, chem-ical and combustion properties of blended fuel should be similar to (marine) diesel.

2. Methodology

2.1. Experimental setups

The combustion test rig used in this work is a commercially available facility from Fueltech Solutions, commonly reffered to as the combus-tion research unit (CRU). This device is based on the well-established constant volume combustion chamber technology. It decouples the ambient temperature from the pressure, which is more challenging for reciprocating engines. In this way, the influence of process parameters such as chamber temperature, pressure, fuel pressure, and injection strategy can be isolated and individually validated. Additionally, the combustion properties of different fuels can be compared under the same operation due to the stable operating conditions. As is shown in Fig. 1, synthetic air and nitrogen supplied from externally pressurized gas bottles are let into the combustion chamber via the plug in the bottom. In this way, chamber pressure can be maintained and the oxy-gen concentration can be manipulated by regulating the ratio of the two gases. The chamber temperature is controlled by the heating device located around the chamber wall and the test isn't started until the wall temperature is within ± 1 °C of the set temperature for 150 seconds. A commercially available injector provided by Bosch (Type: CRIP2) en-ables the direct injection of tested fuel into the chamber, which can be adapted to the specific research requirements. Specifically, this direct injector has an umbrella angle of 158°, sac volume of 0.23 mm³, and 7 nozzle holes (nozzle hole diameter: 0.16 mm). Bboth single and multiple injection strategies are possible depending on the specific research focus. As is shown in Table 1, fuel pressure can be boosted up to 1500 bar

Table 2

Major properties of tested fuels.

Tested Fuel	Cetane number	LHV [MJ/ kg]	Flashpoint [°C]	Density [kg/m³]	Viscosity [cSt]
HVO[37]	79	44	83	778.7 @60 °C	2.82 @60 °C
MGO[38]	47	42.7	60	855 @15 °C	2.6 @40 °C
FAME [39]	45–72.7	37.1–40.4	96–188	855–900	3.89–7.9
EN590 [37]	56.5	43	59	832.4 @60 °C	2.56 @40 °C
HPOfpbo	≥35	43.3	<12	834	1.48 @40 °C
HPOfpboPT	≥35	44.3	64	877	2.5 @40 °C
HPOmisc	≥35	42.9	13	86.6	1.85 @40 °C

and total injection duration is limited to 1.5 ms. The detailed informa-tion and operating speicificaitons of CRU can be found in [33].

The dynamic viscosity of all the fuelswas measured at different temperatures by an Anton Paar Rheometer MCR 302. Different tools are available depending on the value of the viscosity. Highly viscous samples use a plate and cone technique whereas low viscosity samples are measured by a Couette type bob and cup viscometer (double gap method). In this work, a double-gap coaxial cylinder system is used to determine the viscosity of fuels. The double-gap coaxial cylinder system includes a fixed cup containing the liquid sample and a rotating bob. Two connected thin cylindrical annuli of the liquid sample were used to provide shear stress. More detailed description and operating about this setup can be found in [34].

2.2. Definitions

The time-resolved pressure trace in the combustion chamber, as is shown in Fig. 2, is automatically recorded for 47 ms and saved by the software control system after injection. The start of combustion (SOC) is defined as the time when a 0.2 bar pressure rise above the pre-set chamber pressure is reached. This is consistent with the definition in the ASTM D7668-17 standard. While the end of combustion (EOC) is set at the moment of 95% maximum pressure. The ignition delay (ID), it is defined as the time interval from the start of injection (SOI) to SOC and the burn duration (BD) is referred to as the period between SOC and EOC. The rate of heat release (ROHR) can be calculated by Equation 1 based on the first law of thermodynamics, where V is the volume of the combustion chamber and dp/dt is the pressure rise rate calculated from chamber pressure (per 0.02 ms). The specific heat ratio γ can be esti-mated from the average composition and T using the NASA polynomials. However, in the constant volume setup, the ROHR is nearly proportional to the pressure rise rate (in fact the pre-factor $1/(\gamma-1)*V$ varies only slightly since the total pressure rise and thus average temperature rise is rather limited). Therefore, a ROHR is not included and only the pressure rise rate profiles are shown in the paper. More details about the defi-nition can be found in [35].

$$\text{ROHR} = 1/(\gamma-1)*V*dp/dt \quad (1)$$

2.3. Tested fuels

In this work, the hydrotreated pyrolysis oil (HPO) supplied by BTG [36] is produced from fast pyrolysis bio-oil (FPBO) by catalytic hydro-treatment at elevated pressure. The FPBO is produced from thermal cracking of biomass feedstock in the absence of oxygen at 450–500 °C. The liquid yield of this process is around 60–75 wt%. The degree of upgrading is varied and will finally be determined by the requirements for a drop-in fuel and the blend ratio. The quality of the fuel can be controlled by adjusting the hydrotreatment process conditions

Fig. 3. HVO/HPO fuel blends samples from 0 to 100 vol% blend ratio.

(temperature, H2 pressure, residence time) and the catalyst used. The pyrolysis oil itself can be produced from various biomass streams (e.g., wood residues, roadside grass, etc). In this work, five different HPO are being used. They are termed as following: regular HPO upgraded from FPBO (HPOfpbo), HPO derived from miscanthus (HPOMisc425), HPO derived from lignin (HPOLign425), and post-treated HPO to obtain a higher flash point (HPOfpboPT, HPOLign425PT). For benchmarking, commercially available fossil fuel (EN590, MGO) and biofuels including hydrotreated vegetable oil (HVO), fatty acid methyl ester (FAME), are also tested for the same operating conditions. The major chemical and physical properties of tested fuels are listed in Table 2, where the properties of HPOs were measured by BTG according to in-house method.

2.4. Experimental procedures

In the first part of this investigation, both commercially available fuels (MGO, HVO, FAME, and EN590) and HPO fuels were tested at 30 bar chamber pressure and various chamber temperature (430 ~ 580 °C) without EGR. Then, HPO fuels are blended with HVO from 0 vol% to 100 vol% in steps of 25% and tested under the same operating condi-tions. Fuel pressure was maintained as 1500 bar and a constant injection duration time of 1.5 ms was applied during the tests for all fuels. Be-tween tests, old fuel is bled off manually and the fueling system is flushed by the new fuel sample when to be tested. As a standard routine, up to 13 injections are performed to further ensure no old fuel is left in

the injection system. Only 8 injections are conducted in each measure-ment in a formal test, where the first three trial injections are dismissed and only the last 5 injections are automatically recorded in the CRU software. The results presented in Section 4 are the assembled average of these 5 injections for each measurement. An error bar is not included in the pressure and pressure rise rate profiles for the sake of clarity. Nevertheless, for EN590 and MGO the repeatability is explicitly tested. The results shown in Fig. 11 in the Appendix indicate very good repeatability of the measurements.

The tested HVO/HPO fuel samples (note that HPOfpbo is used to make the blends) are shown in Fig. 3. Noteworthy is that they were stored for more than 30 days without showing any visible phase separation. The viscosity of fuels was tested from 10 to 100 °C in steps of 15 °C. An equilibrium time of 15 min was used for each temperature, after which the dynamic viscosity of the sample was consecutively measured 10 times with a time interval of 10 s and is averaged to obtain the final dynamic viscosity result. The difference between the 10 consecutive measurement results is negligible, hence the error bar was omitted in the results.

3.   Results   and   discussions

3.1. HPO benchmarking

Fig. 4a displays the ID time of the tested fuels at 30 bar and various chamber temperatures. Basically, the ID time can be categorized into three groups: the HPO fuels, diesel-like fuels (EN590, FAME, and MGO), and HVO. To be more specific, the HPO fuels display the longest ID, while diesel-like fuels present similar IDs among each other. HVO yields the shortest ID. This is in good agreement with the cetane number of the tested fuels listed in Table 2. It also can be noticed that the ID of all fuels decreases as the temperature increases, which is expected since both the physical and chemical delay decrease. In addition, the ID differences among different fuels present a decreasing trend at high temperatures. As is illustrated in Fig. 12a in the Appendix, the relative ID difference among tested fuel compared to HVO decrreases. This is beneficial when it comes to real engine apllication at high load, where ambient tem-perature is always quite high. So that HPO can auto-ignite more like commericial fossil fuels. Furthermore, the HPO derived from different origins also show small differences in ID at the tested condition. The ones with post-treatment (PT) to increase the flash point display shorter ignition delay. For the burn duration (BD), the categorization in the three groups is not that obvious. For instance, HPO425PT and FAME show
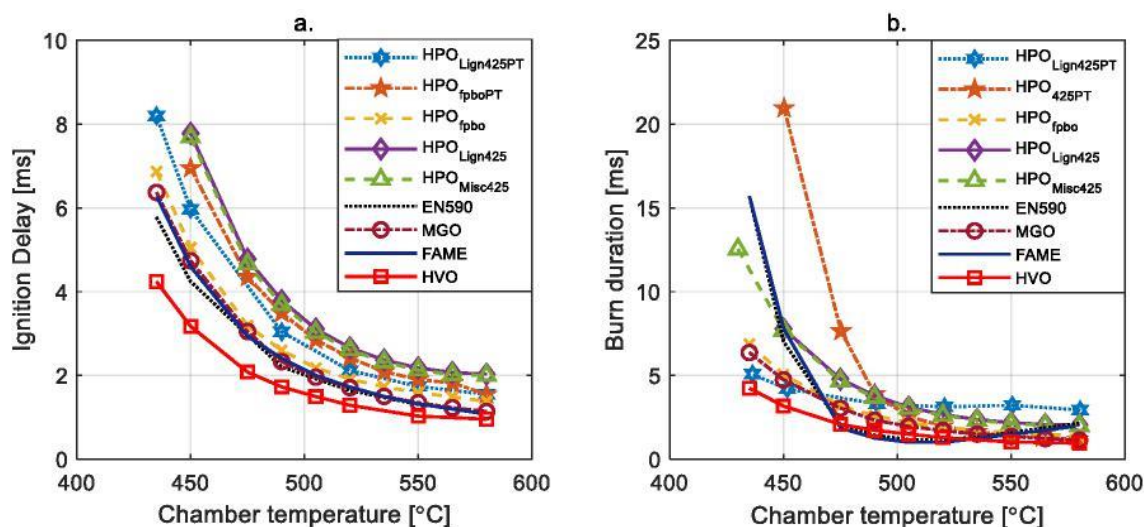

Fig. 4. Ignition delay (a) and burn duration (b) of tested fuels at various chamber temperatures.
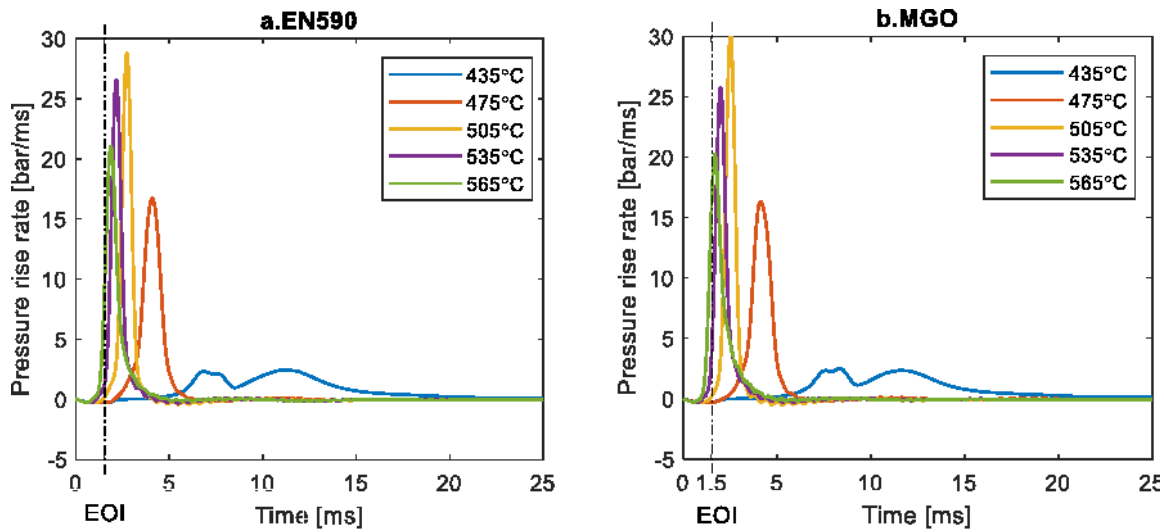
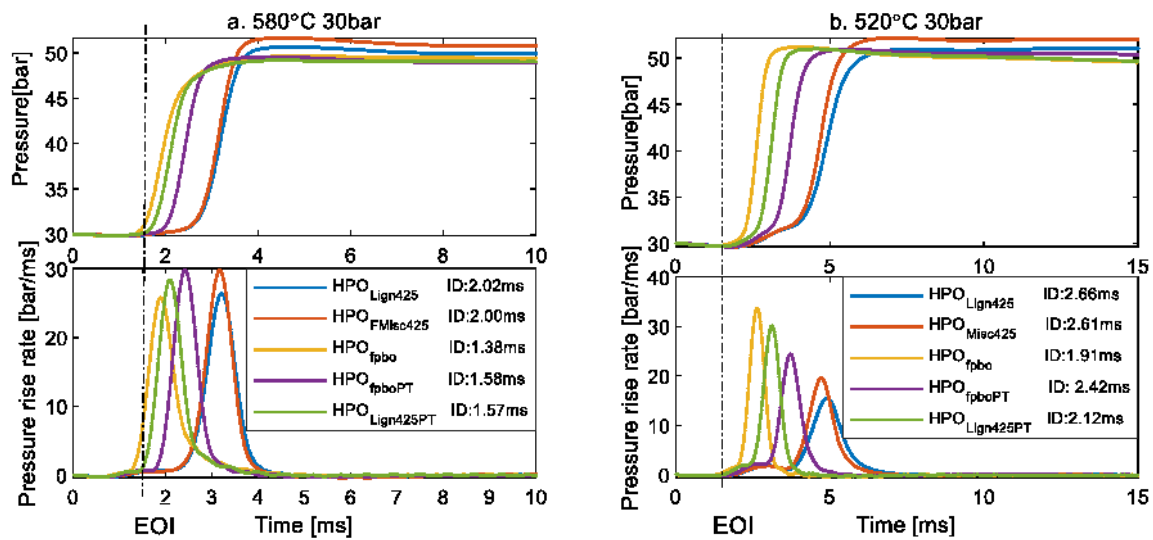**Fig. 5.** Pressure rise rate of EN590 and MGO.

Fig. 6. Pressure rise rate and chamber pressure of tested HPO fuels.

a much longer BD at low temperatures than EN590 or HPO425lignPT for that matter. Generally BD decreases when the chamber temperature increases for all (bio) fuels as is shown in Fig. 4b. A transition temper-ature seems to exist for the fossil fuels (MGO and EN590), afterw which the BD starts to increase again. As is shown in Fig. 4b, BD of MGO and EN590 increases slightly after 505 ˚C. Since the increment is small, validation tests were done to investigate whether this was systematic or random. These showed identical results, as shown in Fig. 11 in the Appendix. This is mainly because of the transition of two combustion regimes illustrated by pressure rise rate profiles in Fig. 5. It can be clearly noticed that at 435 ˚C, both the ID of EN590 and MGO is long due to low ambient temperature. A long separation between the end of injection (EOI) and SOC is shown, and the heat release process is quite slow. Both EN590 and MGO clearly illustrate a two-stage heat release mode in their PRR profiles. This is most probably due to the fact that the fuel has mixed readily with the ambient before ignition, approaching a homog-enous (lean) mixture purely governed by chemical kinetics. As the chamber temperature gradually increased to 505 ˚C, low temperature heat release (LTHR) peak becomes invisible and high temperature heat release (HTHR) peak increases remarkably. Still, the separation among

injection and combustion events exists and a premixed-dominant com-bustion process is displayed which is still mostly governed by the chemical kinetics. Consequently, the heat release gets faster and BD decreases. However as the temperature increases beyond 505 ˚C, ID further decreases, and heat release starts before the EOI, and the injec-tion and combustion overlap. Therefore, the mixing time decreases and burn duration is not only dominated by kinetic effects but also by the length of the injection (note that combustion cannot end before injection has stopped). Concequently the BD increases again as the chamber temperature increases.

The above-mentioned two combustion regimes not only depend on the ambient temperature but also the reactivity of the fuel itself. Fig. 6 shows the heat release process of all tested HPO fuels at two typical temperatures. At 580 ˚C, the ID of all HPO is relatively short. It can be seen that pressure rise starts before the end of injection (EOI), the combustion consists of mixing-controlled combustion and premixed combustion. The longer ID cases generally show a higher and later PRR peak due to extended mixing. Hence the mixture more and more become less stratified. At 520 ˚C, injection finishes before combustion for all HPO's, leading to more time to mix before combustion. In fact the two-
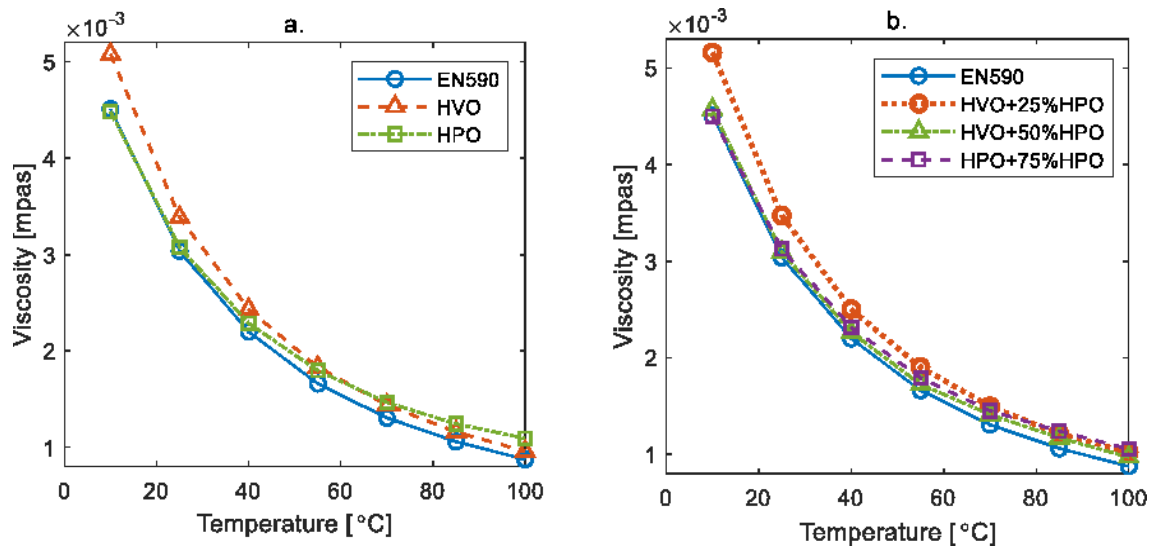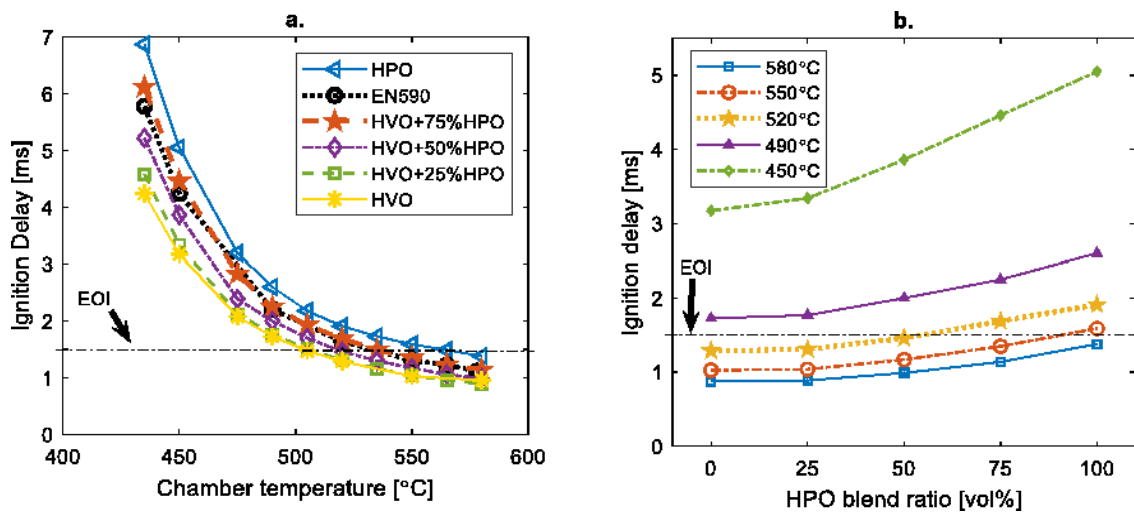
Fig. 7. Viscosity of tested fuels.

Fig. 8. Ignition delay as a function of chamber temperature and HPO blend ratio.

stage heat release, as can be seen in Fig. 6b, becomes more prominent strongly correlating with the actual ID at these conditions.

3.2. HPO as a drop-in fuel

The ultimate goal of this work is to investigate the potential of bio-fuel in HD trucks and marine applications. Therefore it would be ideal that the HPO has identical properties as commercially available fossil fuels (for example: diesel) so that the engine requires minor/no modi-fications. This section compares the viscosities and combustion prop-erties of HVO + HPO fuel blends and EN590. As is shown in Fig. 7a, the viscosity and its dependency on temperature of HPO, EN590, and HVO are nearly the same. For the blends, interestingly, no clear correlation between blend ratio and the viscosity can be noticed. The difference in viscosity among different blend ratio cases is minor. For example, HVO + 25%HPO shows the highest viscosity when the fuel temperature is below 55 ℃, a maximum 7.6% difference is shown at 10 ℃ in Fig. 7b。

Fig. 8 presents the ignition behavior of neat EN590, HVO, HPO, and HVO + HPO blends. It can be observed from Fig. 8a that ID decreases at high chamber temperature. Furthermore, the ID differences relative to HVO generally decreases as the tempereature increases (shown in

Fig. 12b). It indicates that the influence of ambient conditions on igni-tion becomes less crucial at high temperatures. Further to notice, the ID clearly increases when the HPO ratio increases, irrespective of chamber temperature. It indicates that the reactivity of these specific fuel blends can be modified by manipulating the composition of the fuels at each condition. And it is interesting to find that the ID of HVO + 75%HPO is identical to EN590. To make sure these two fuels show a similar overall combustion process, the chamber pressure and pressure rise rate profiles from 475 to 565 ℃ cases are shown in Fig. 9. It can be seen that both the chamber pressure and PRR profiles of HVO + 75%HPO and EN590 are nearly identical at these temperatures, which verifies that these two fuels show similar ignition and combustion behavior. This is important when it comes to real engine applications that the ECU control system would need no modification to the original engine setup. Note that the HVO + 75%HPO fuel blends are nearly 100% CO_2-neutral from a life-cycle point of view as both are derived from renewable resources and align with the REDII directives.

Fig. 10 displays the chamber pressure and pressure rise rate profiles of HVO + HPO blends from 0 to 100% blend ratio. Though all cases show gaussian-like PRR profiles, consisting premixed-combustion phasing and a small fraction of burn-out. Again, two different combustion regimes at
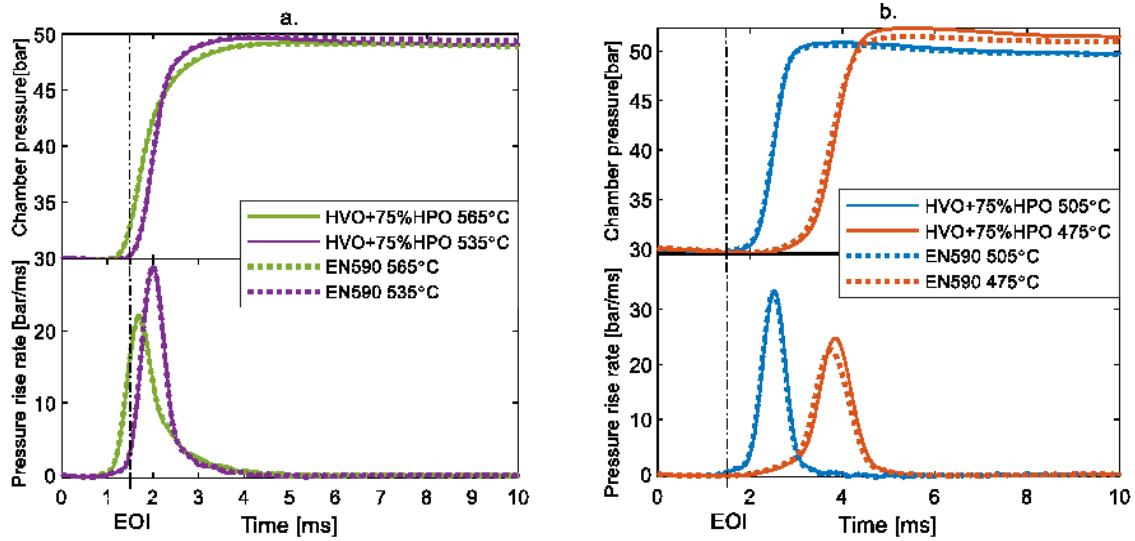
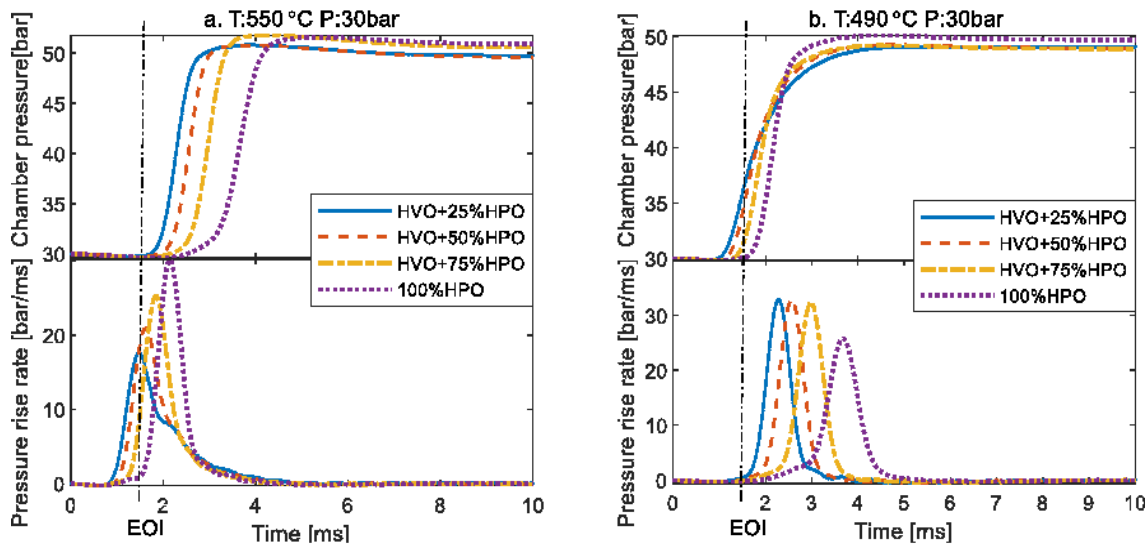**Fig. 9.** Chamber pressure and pressure rise rate of EN590 and HVO + 75%HPO blend.

Fig. 10. Pressure rise rate and a chamber pressure of HVO + HPO fuel blends at various blend ratios.

different temperature are shown. At high-temperature range in Fig. 10a, where the ID is relatively short, combustion starts before the end of the injection event. ID increases as the HPO blend ratio increases. The extended mixing time results in more premixed-dominent combustion and higer peak of PRR. Whereas at lower temperature cases (490 °C), the ID time is relatively longer, combustion starts well after the injection event. The increased HPO blend ratio further extends the mixing time. So that he fuel/air mixture approaches over-mixing state. Consequently, the PRR peak of these cases decreases at these high HPO blend ratio cases, as is shown in Fig. 10b.

4. Conclusions

1. Based on the operating conditions, the ignition delay time of tested fuels is ranked according to HPO > diesel-like fuels > HVO. It is also revealed that the biomass origin of HPO has a minor influence on the ignition delay. And this influence decreases at high chamber temperature, which applies to both the absolute and relative ID. More-over, it is shown that post-treatment increases the ID of HPO fuels.

2. HVO and HPO fuel blends show good stability. The viscosity of all tested fuels decreases as temperature increases as expected. The differences in viscosity among HVO, HPO, and EN590 are small. No clear correlation can be observed between the viscosity of HVO + HPO blends and the blend ratio.

3. Blending the HPO with HVO can substantially increase the ignition delay time of HVO and the ignition delay time is proportional to the blend ratio. Two combustion regimes are observed at different temperature ranges. They mainly depend on fuel reactivity. Up to a certain ID-EOI, PRR peak increases as the temperature increases due to the increase of the premixed combustion part in the classical diesel combustion. At a certain difference, the PRR peak decreases due to over-mixing when the mixture is overall lean.

4. At 75 vol% HVO blend ratio, HVO + 75 vol%HPO have an identical ignition delay time as EN590 nearly independent of the operating conditions. Even more, both the chamber pressure and PRR profiles are nearly identical as well. This indicates a fully CO2-neutral biofuel can be created that behaves exactly the same in a compression ignition engine.
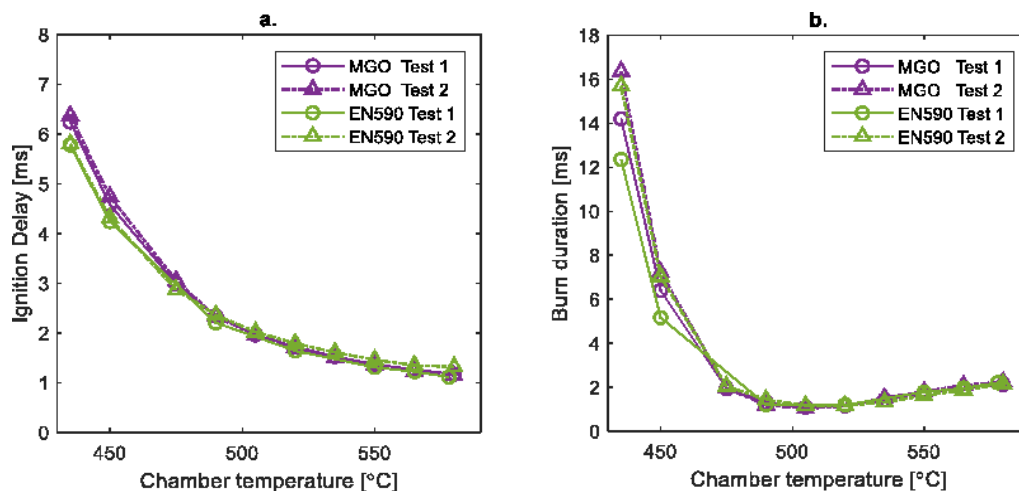
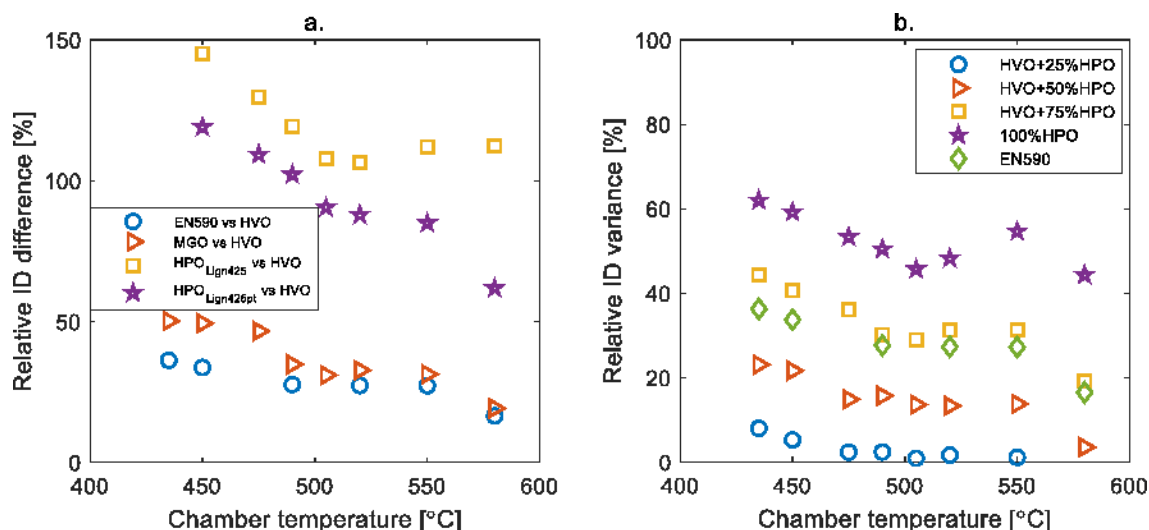**Fig. 11.** Ignition delay and burn duration of MGO and EN590 in different test.

Fig. 12. Relative Ignition delay difference between tested fuels and HVO.

CRediT authorship contribution statement

Jinlin Han: . **L.M.T. Somers:** Formal analysis, Resources, Supervi-sion, Writing – review & editing. **Bert van de Beld:** .

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix

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