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Comparative analysis of waste-derived pyrolytic fuels applied in a contemporary compression ignition engine

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The outcomes of research regarding pyrolysis oils obtained from waste sources (WPO) used to power a compression-ignition engine have been presented in this paper. Oils obtained in an industrial process based on polypropylene (PPO), polystyrene (PSO) and used car tires (TPO) were used. Prior to conducting engine tests, a in-depth examination of the tested fuels parameters was undertaken. An advanced single-cylinder research engine utilizing split fuel injection technique was used for the tests. Emission analysis was performed using multi-compound FTIR analytical system. The WPO were blended with diesel fuel in proportions of 20%, on the mass basis and tested at middle engine load and variable EGR rates. Tests have shown that modern combustion systems compliant with the Tier 4/Stage IV standard with multi-pulse injection can handle fuels with a WPO content of 20% without the need for recalibration. The addition of PPO did not significantly affect the emission, while mixing with PSO resulted in elevated levels of hydrocarbon and carbon monoxide emissions. Regarding to the mixture with TPO, increased levels of particulate matter, sulfur oxides, aromatic compounds and formic acid were observed.

Key words: *waste, pyrolysis oil, fuel blending, diesel engine combustion, exhaust*

1. Introduction

Worldwide economic growth led to a significant rise in the demand for energy resources. Among these resources, petroleum products continue to play a pivotal role, serving as the primary source of propulsion in the ever-expanding transportation sector [7]. As a result of ongoing electrification, a slight decrease in gasoline consumption is estimated, while the consumption of diesel fuel (DF) increases [3]. The continuing interest in compression-ignition engines results primarily from the fact that they are used in the long-distance heavy transport sector, maritime transport, and off-road applications. The specificity of these sectors significantly limits their electrification, while the development of diesel engine technology has allowed them to consolidate their position. All forecasts indicate that in the foreseeable future, combustion engines will remain the main driving force in the transport sector [6].

The aspirations to meet the global warming targets outlined in the 2015 Paris Agreement are driving the expansion of the renewable energy sector. Renewable fuels are estimated to contribute about 15% of primary energy production by 2040 [7]. Nevertheless, the same projections suggest that this will only suffice to meet half of the rising energy demand. It is therefore reasonable to intensify research in the area of fuels from renewable sources. Currently, two varieties of renewable diesel, fatty acid methyl esters (FAME) and hydrotreated vegetable oil (HVO), present a high level of technological feasibility, allowing production on an industrial scale. However, some specific properties make it difficult to use mentioned fuels independently to power an internal combustion engine [22]. While their use in a mixture with conventional DF is possible to a limited extent. A significant disadvantage for both FAME and HVO, in the case of first generation fuels, is the necessity to compete for raw material with the food industry [26].

The solution to the above issues is the introduction of feedstock diversification in the production of alternative fuels. To ensure continuity and profitability of production, the raw material should be easily available in large quantities and affordable. An example of such raw material are the end-of-life tires (ELT) [20]. Approximately seventeen million tons of them is produced worldwide annually [21]. ELTs are characterized by huge availability [24], but from the waste management side it is a cumbersome raw material, difficult to store and process [26]. Among the available ELT recycling technologies, pyrolysis process deserves special attention.

Pyrolysis is a technology that perfectly fits into the strategy of sustainable development. The process encompasses the thermal decomposition of solids within an environment devoid of oxygen [2]. It allows simultaneous synthesis of both synthetic and biological materials [9]. It provides a waste management possibility that is unavailable to other recycling methods. Referring to recent studies [10], the tire pyrolysis process is divided into four stages depending on the temperature.

The process parameters and reactor type are selected to maximize the yield of the desired fraction. Regardless of them we get rid of problematic waste and obtain (33–39%) weight char, (34–55%) weight oil, and pyrolytic gases [8, 25]. The high calorific value of pyrolysis gases, reaching approximately 30–40 MJ/kg, enable the execution of the pyrolysis process, making it self-sufficient [5, 8].

Obtained oil, called tire pyrolysis oil (TPO) is dark brown fluid exhibiting properties akin to diesel oil and gasoline [4, 18].

Nevertheless, this prospective fuel possesses several characteristics that hinder its direct utilization in an internal combustion engine. The sulphur content is considerably higher compared to DF [17]. Raw TPO contains it from 0.55 to 3.95% [11]. The presence of heteroatomic molecules and the elevated concentration of aromatic compounds are deemed unacceptable considering environmental protection and necessitate the refinement of the fuel.

Another example of waste materials, suitable for the pyrolysis, which do not contain sulphur are plastics. It is worth emphasizing that the stream of waste generated in Poland and in the world has remained at a constant level for many years. In Europe, collect of the post-consumer plastic waste amounted to 27.3 million tonnes in the year 2016 [14]. This allows to treated plastic waste as a continuous source of energy that can be used in fuels' production. Analysis of the morphology of mixed plastic waste indicates the following percentage distribution of different types of plastics in the overall waste composition: polyethylene terephthalate (PET): 25%, polymer films: 19%, polypropylene (PP): 11%, high-density polyethylene (HDPE): 8%, polystyrene (PS): 4%, other types of plastics: 3% [19].

The advantage of plastic waste is its high amount of energy, confirmed by the elevated value of the heat of combustion. The main thermochemical methods used during waste conversion include gasification, pyrolysis, and combustion. The latest research of Szwaja et al. [23] confirmed high combustion process similarity between conventional DF and pyrolytic oils. However, pyrolysis allows the production of a liquid fraction with properties similar to conventional fuel used today [1]. The pyrolytic oils from plastic feedstock have already been thoroughly researched in terms of processing technologies and fuel properties; however, feasibility tests in compression ignition engines are scarce, especially if modern engine platforms are considered.

Even partial substitution of fossils with waste pyrolytic oils (WPO) may turn out beneficial considering factors such as depletion of fossil fuel reserves, and overall emissions in the well-to-wheel approach (even considering elevated tailpipe emissions). Reasonable waste disposal may give additional value to the entire process.

Given that researchers employed diverse engines and various pyrolysis oils, comparing the results becomes challenging. Nevertheless, some overall picture of the WPOs as diesel engine fuels appear based on the available studies. When compared to diesel, most of the WPOs exhibit lower cetane numbers and higher amounts of unsaturated hydrocarbons. It can have a detrimental impact on exhaust emissions and decrease engine thermal efficiency [16]. Hence, in the majority of studies, a combination of pyrolytic oil and diesel fuel blends was utilized [16].

The above introduction clearly outlines the significant potential of WPO fuels. Nevertheless, an extensive literature review by Mikulski et al. [15] clearly revealed knowledge gaps regarding the use of them. This study aims to presenting a thorough comparative analysis of the most popular WPO varieties. The WPO used in this study was obtained through a process optimized specifically for engine fuel applications. To enhance our understanding of the combustion process, the study determined the key physicochemical parameters of the obtained fuels. Engine tests were conducted using a cutting-edge testing platform, with consistent testing procedures applied to each fuel. The outcomes of this research offer valuable and unprecedented comparative data, guiding future investigations into the potential of WPO fuels.

2. Materials and methods

2.1. Pyrolysis oils used in the experiment

The research involved pyrolysis oils derived from two distinct input materials: used tires and plastic.

The TPO was obtained through an industrial process in which the reactor was fed with used tires without any preprocessing. In contrast, the plastic pyrolysis oil was produced from a homogeneous material on a laboratory scale. Specifically, commercial granulated polypropylene (PP) and polystyrene (PS) with granulation of 2 × 4 mm were used as the input materials. The acquired oils underwent a two-step filtration process, involving a Buchner funnel with a 13 mm mesh on the fine-filter side. Process of obtaining the tested oils has been extensively discussed in another work by Januszewicz et al. [12].

The derived oils underwent both physical and chemical analysis. The gas chromatography and mass spectrometry (GC-MS) technique was employed to ascertain the detailed molecular structure of them. In the authors' previous research can be found a detailed description of fuel testing methods, for TPO [16] and for plastic pyrolysis oils [12]. The key properties of the examined fuels were collected in Table 1.

Table 1. Physical and chemical parameters of the fuels

Parameter	DF	TPO	PPO	PSO
Density @ 15°C [kg/m^3]	826	948	776	942
Viscosity @ 40 °C [mm^2/s]	2.3	3.7	1.69	1.22
Flashpoint [°C]	61	90	< 24	34
Sulphur content [mg/kg]	6.1	5000	–	–
Water content [mg/kg]	11	410	–	–
Higher heating value [MJ/kg]	44.8	44.7	44.7	40.5
Cetane number	55.3	22.3	27.5	–

Many of the mentioned properties indicate the necessity of blending WPO and DF. It is undertaken to guarantee the secure and efficient working of the engine when using pyrolysis oils. Consequently, a decision was made to create mixtures consisting of pyrolysis oil in 20% proportions, combined with the suitable quantities of EN 590:2004/ASTM D975-compliant automotive diesel without any bio components. The formulation of these blends was determined by the mass ratios of the individual components. The crude TPO underwent distillation due to its elevated viscosity and low flashpoint. Further research was conducted on the fraction obtained within the temperature range of 330–375°C, designated as TPO F4.

The mixtures of TPO F4 and DF were labelled as TPO. A similar naming convention was applied to label the mixtures of polypropylene oil (PPO) and polystyrene oil (PSO) with DF.

2.2. Research engine test stand

The experimental work encompassed a comprehensive analysis of an engine operation at Lublin University of Technology. The research focused on an advanced single-cylinder AVL 5402 research engine, as detailed in Table 2.

Table 2. Research engine specifications

Type	AVL 5402
Configuration	four-stroke, single-cylinder
Bore/stroke	85/90 mm
Displacement	510.5 cm^3
Compression ratio	17:1
Swirl ratio	1.7:1
Combustion chamber	Mexican hat, toroidal type
Injection system	CR, Bosch CP4.1; 180 MPa; multi-pulse capability
Intake path	Electrically driven Eaton M45 compressor; with thermal management
EGR system	high-pressure, with thermal management
Engine management	AVL-RPEMS, ETK7-Bosch
IVO/IVC	712/226 CAD
EVO/EVC	488/018 CAD

In order to replicate real operating conditions, the AVL asynchronous motor dynamometer with speed control was connected to the engine. Figure 1 illustrates the test stand setup.

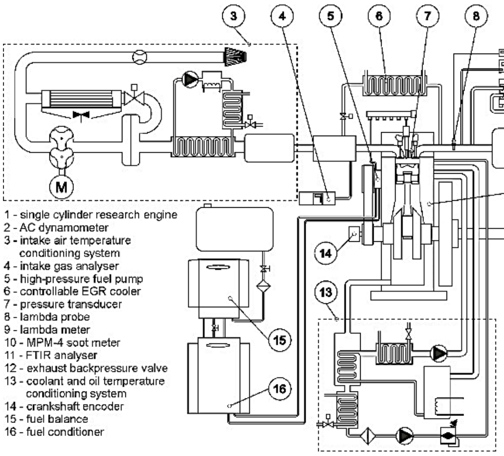

Fig. 1. Diagram of the engine test bench

The combustion system featured a four-valve head and a toroidal in-piston combustion chamber. To condition the fuel, the AVL 753C temperature conditioner and AVL 733 S dynamic fuel meter were employed. Fuel was introduced into the combustion chamber through a Bosch CP 4.1 high-pressure pump, employing a seven-hole electromagnetic injector featuring a 145° spray angle. The injection parameters were managed through ETAS INCA software and a fully open Bosch controller.

Electrically powered Roots compressor Eaton M45 supplied the high pressure. A proprietary thermal conditioning system was implemented to sustain consistent temperatures for coolant, lubricant, and charge air, with an accuracy level within ±0.5°C.

In order to evaluate the excess air ratio and account for compensation of the pressure, as discussed in reference [16], the experiment employed a Bosch LSU 4.2 lambda probe and an ETAS LA4 lambda meter.

The concentrations of 23 exhaust gas components, both regulated and non-regulated, were monitored using an AVL FTIR multi-component analytical system. Additionally, particulate concentration was measured by a Maha MPM-4 analyzer.

For combustion analysis, an AVL GU22C piezoelectric pressure transducer was installed directly in the engine head. The initiation of the pressure signal recording was carried out by an optical encoder, ensuring a consistent angular resolution 0.1 CAD.

2.3. Research engine matrix

The research was performed at a single operating point, at IMEP set at 0.5 MPa, and at a constant engine speed of 1500 rpm. This aligns with a specific point in the previous tests, which significantly contributes to the overall emissions. Injection timings (SOI), and the EGR ratio were varied to examine the engine's response to variable control parameters when fueled with WPO in comparison to DF. Four distinct levels of EGR were investigated for each tested SOI.

Experiments were conducted under controlled thermal conditions, with the engine coolant, lubricating oil, and recirculated exhaust gas at the manifold inlet maintained at a temperature of 85°C. The intake air temperature was held constant at 35°C. The final aspirated charge temperature was determined by the enthalpy balance between fresh air and EGR, and was left uncontrolled, typical conditions encountered during standard engine operation. The temperature of the fuel supplied to the high-pressure pump was consistently held at 30°C. The engine calibration was meticulously tailored for the standard diesel fuel, with the primary goal of minimizing overall emissions while preserving a high indicated thermal efficiency. Details regarding the engine calibration for all operating points can be found in Table 3.

Table 3. Engine operating conditions and control parameters.

IMEP [MPa]	0.5				
MAP [MPa]	0.125				
SOI1 [CAD]	336	338	340	342	344
SOI2 [CAD]	350	352	354	356	358
Fuel pressure [MPa]	80				
Pilot fuel quantity [mg]	1.6				

The testing procedure initiated with the reference DF, followed by the blended fuels, each following an identical protocol. To ensure the intended ratios of the tested fuels, the entire fuel system underwent thorough draining and multiple flushing cycles with the chosen fuel during each fuel transition. Each blend underwent three repetitions of the test sequence.

At each engine operating point, once all parameters had stabilized, in-cylinder pressure was meticulously recorded for 100 cycles, and slowly changing data were continuously monitored during the 30-second measurement interval. The presented data represents the mean values derived from three separate engine runs for each testing point. For the reference DF, supplementary measurements were conducted to ascertain the precision of emission measurements.

2.4. Data analysis procedures

The combustion analysis, utilizing the AVL Boost software, relied on in-cylinder pressure data. This analysis incorporated gas-flow models, internal EGR estimation, and the computation of heat transfer through the cylinder walls. The latter was determined utilizing the Hohenberg correlation [13]. The heat release rate (HRR) was determined by analyzing the cylinder pressure using a first-law analysis. The HRR values reported in the paper are gross values, which consider the calculated heat transfer rate (Q_{ht}). The cumulative HRR values were then used as the foundation for computing the mass fraction burned (MFB), which, in turn, facilitated the calculation of combustion timing indicators.

To determine the maximum measurement error for directly measured values, we considered either the standard deviation from three samples or the accuracy of the measuring device, depending on which value was greater. For indirectly calculated values, the measurement error was determined using the partial derivatives method, as outlined by Kline and McClintock [13].

For a more in-depth exploration of uncertainty analysis readers are directed to Mikulski et al. [16].

3. Results

3.1. Analysis of combustion

The impact on the combustion of the examined fuels evaluate with reference to the diesel combustion. Figure 2 show in-cylinder pressures and HRR curves at reference conditions. Figure 3 illustrates HRR curves for WPO blends, with DF as a reference. The figure also demonstrates the combustion response to external EGR.

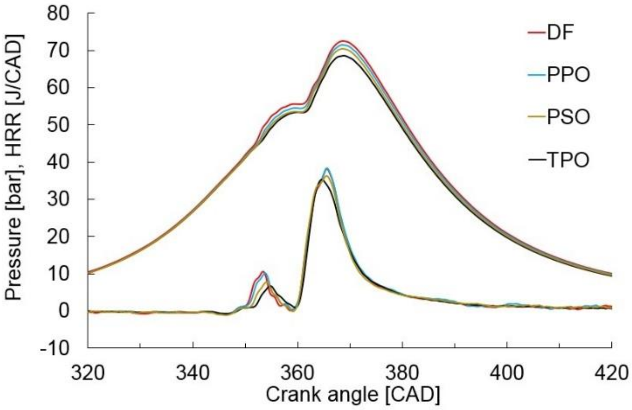

Fig. 2. In-cylinder pressure for all investigated fuels at reference conditions

Figure 2 clearly shows that the addition of WPO at a level of up to 20% does not negatively affect the combustion process. The pressure curves have the shape of a typical diesel combustion characteristic. The intense rate of pressure increase begins at the top dead center (TDC) regardless of the added WPO. After the initial injection of pilot fuel, a minor reduction in HRR occurs due to fuel vaporization. Following this, low-temperature reactions occur, yielding reactive species that do not contribute significantly to heat generation but cause earlier ignition of the pilot fuel spray. The initial prominent peaks observed in the HRR curves stem from the high temperature heat release associated with the predominantly premixed pilot fuel. At 354 CAD, the injection of the main fuel commences. The auto-ignition of the main fuel spray occurs shortly after it enters the heated region of the pilot combustion. When examining the principal HRR peak, one can identify the shift from kinetic to mixing-controlled combustion. This transition occurs at roughly 363 CAD for all cases illustrated in Fig. 2. The final phase, denoted as the afterburning period, commences at 375 CAD, marked by declining (HRR) values, and maintains consistent across for all fuel blends.

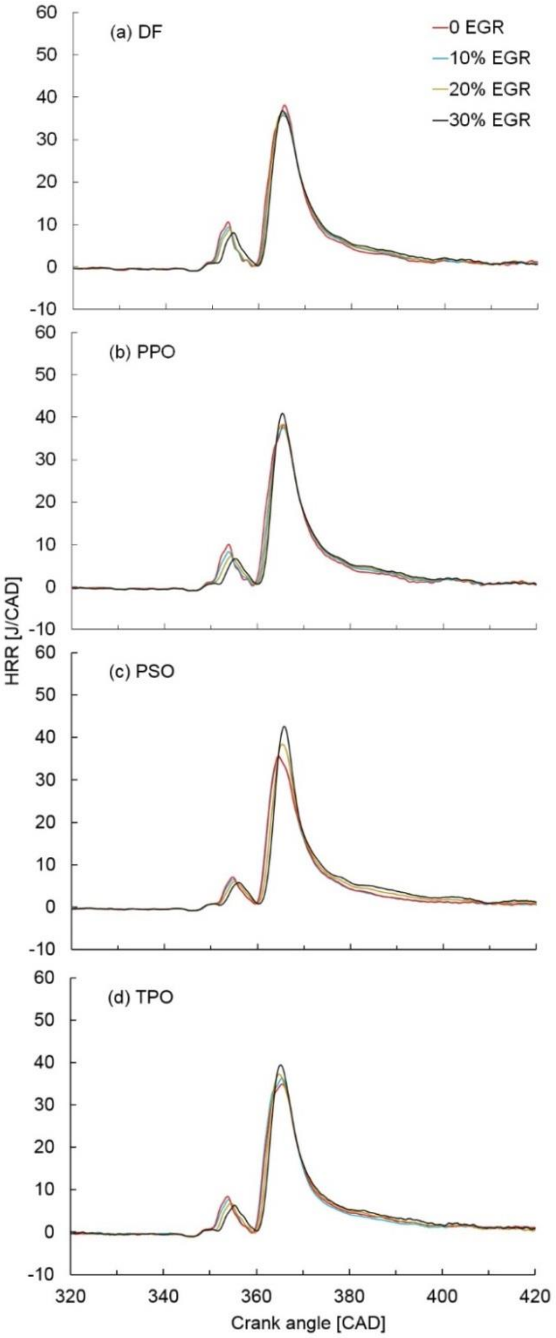

Fig. 3. EGR effects on HRR for tested fuels; 0.5 MPa IMEP, reference SOI

Regardless of the used fuel combustion reaches completion at around 410–420 CAD.

The observed delay in pilot fuel ignition doesn't influence the initiation of main fuel combustion but does result in a somewhat larger amount of unburnt fuel as combustion transitions to the mixing-controlled phase.

The impact of PPO on combustion is negligible compared to the same admixture of TPO or PSO. In the case of TPO, the delay of approximately 3 CAD is due to the much lower cetane number and lower volatility. The most significant influence of the pilot fuel dosage was noted in the case of the PSO combustion process. This is because PPO consists primarily of aromatic hydrocarbons characterized by comparatively low auto-ignition properties.

From Fig. 3, it is clear that the EGR introduction inhibits the premixed combustion of the pilot fuel. The volumetric, kinetic combustion is highly sensitive to the EGR rate, with a proportional delay of approximately 1 crank angle degree for every 10% increase in EGR. This delay results in less fuel being burned before the main fuel injection ignites, which reduces the heat release rate in this stage of combustion. This energy is subsequently discharged during the combustion of the main charge, leading to higher HRR peaks and longer combustion times. This is especially noticeable in the case of TPO combustion.

3.2. Analysis of emissions

As already mentioned, the authors of this work have shown in previous studies that the impact on emissions of DF blends with the addition of WPO up to 20% can be considered negligible in the case of heavy and medium loads. Therefore, in this study, the case of low load is considered. In Fig. 4 were compared, indicated specific emissions of both, regulated and non-regulated exhaust compounds under reference conditions for all tested fuels.

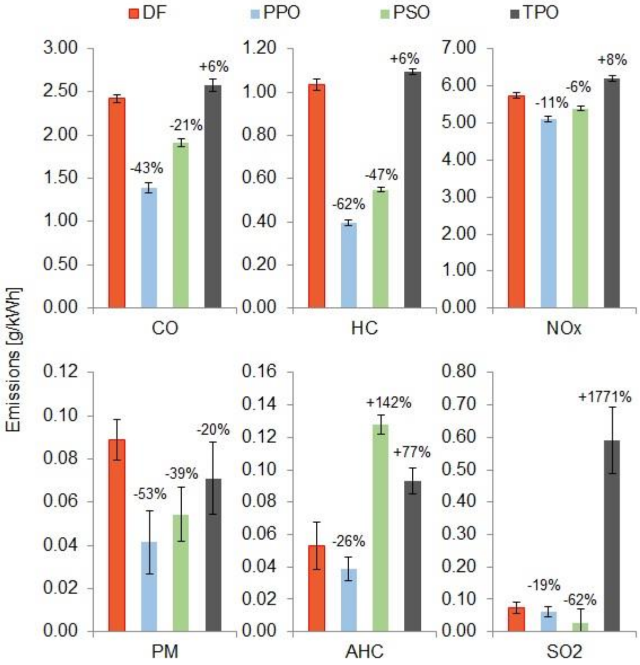

Fig. 4. Indicated specific emissions of regulated and non-regulated exhaust compounds at reference conditions

In this load regime, PPO and PSO samples cause improvement the regulated emissions. The elevated CO levels arise due to the delayed combustion of TPO, extending into the expansion phase and consequently experiencing flame extinction as the piston approaches the opening of the exhaust valve. The rise in NO_x levels for TPO blends is attributed to the CI reduction, causing a shift in combustion to the main high-temperature stage. Increased temperature values during TPO combustion straight result in increased NO_x formation. Considering the measurement uncertainty, the incorporation of a 20% TPO blend does not exhibit any significant impact on particulate matter (PM) emissions.

The exceptionally high emission of aromatic hydrocarbons when using PSO correlates perfectly with the chemical composition of this fuel. PSO contains up to 98% of aromatic compounds.

A similar trend can be observed when using TPO. The high sulfur content in this fuel results in a sharp increase in sulfur dioxide emissions. It should be noted that PPO and PSO fuels do not contain sulfur. The SO_2 emissions recorded during the combustion of these mixtures are solely due to the sulfur contained in the DF. However, the impact of the addition of PPO and PSO is very positive in reducing SO_2 emissions, by 19 and 62%, respectively.

In the ensuing section of the investigation, a PPCI (Partially Premixed Compression Ignition) strategy was employed, using EGR and SOI as parameters to manage emissions, with the goal of achieving a favorable trade-off between NO_x and PM emissions for the various WPO blends tested. The outcomes for PM and NO_x emissions are depicted in Fig. 5 and Fig. 7, showing absolute values for the reference DF fuel. Moreover, for the WPO blends, the differences in PM and NO_x emission values between each specific fuel and DF are illustrated in Fig. 6 and Fig. 8, respectively.

The EGR trade-off between PM and NO_x is evident when comparing the results for the reference fuel DF in Fig. 5 and 7. The minimal PM emissions are attained at lower EGR rates, while the lowest NO_x values occur on the opposite side of the calibration map (note the inverted EGR axis values on Fig. 7).

Significantly, with the utilization of the highest tested EGR rates and advanced SOI, emissions of both NO_x and PM are lower in comparison to DF baseline value for all fuels tested. This combustion regime corresponds to a greater amount of pre-mixing. For the SOI advanced by 4 CAD and 30% EGR, PSO has the capability to achieve a 0.21 g/kWh reduction of PM emission compared to DF. This effect decreases significantly both by decreasing the amount of EGR and by decreasing the injection timing of the pilot dose. We observe similar trends for each blends, but for the PPO and PSO the gain from ignition advance is much more visible than in the case of TPO. The deterioration of the emission rate on the PM side occurs much earlier in the case of TPO, and further delaying the ignition causes an increase in emissions by 0.27 g/kWh compared to DF.

At the same time, the calibration with the lowest particulate content when operating without EGR is accompanied by the highest NO_x emissions, for PPO and TPO. Interestingly, all tested blends showed an improvement in NO_x emission reduction at the highest EGR.

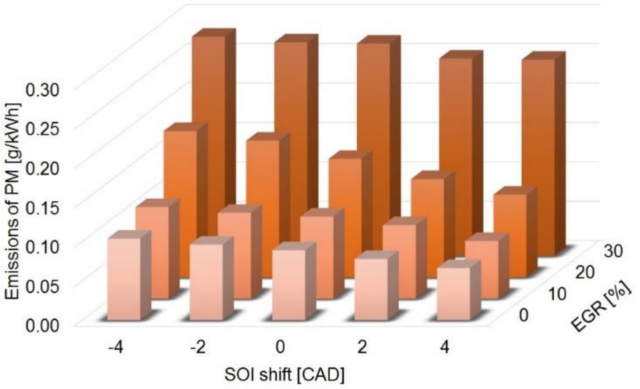

Fig. 5. Indicated specific emissions of PM for DF at variable SOI and EGR rates

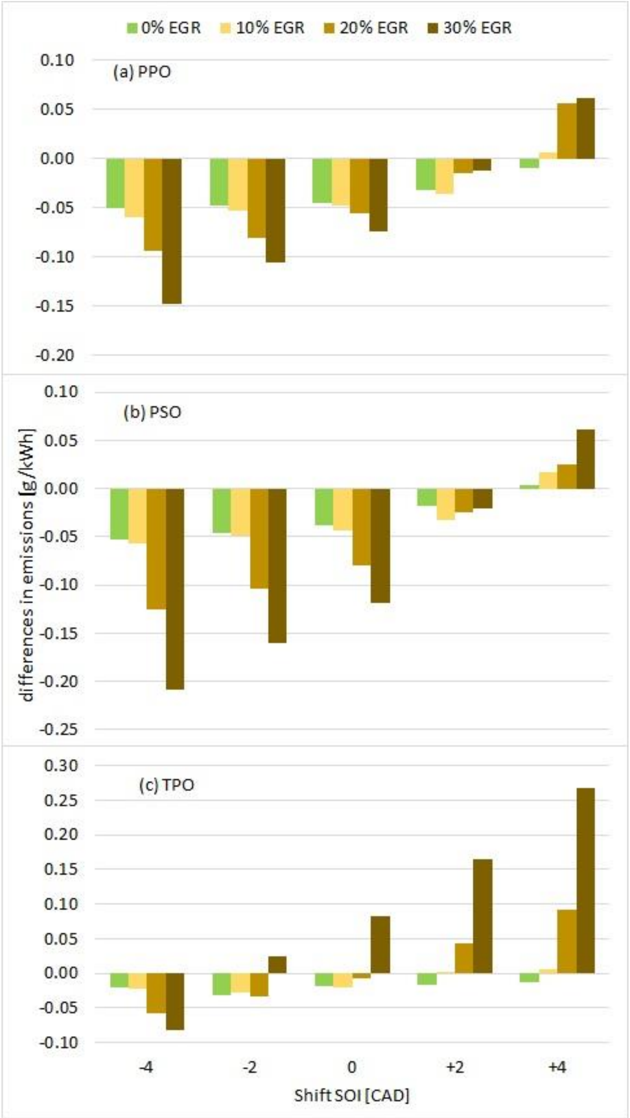

Fig. 6. Differences in PM emissions between DF and tested WPO at variable SOI and EGR rates

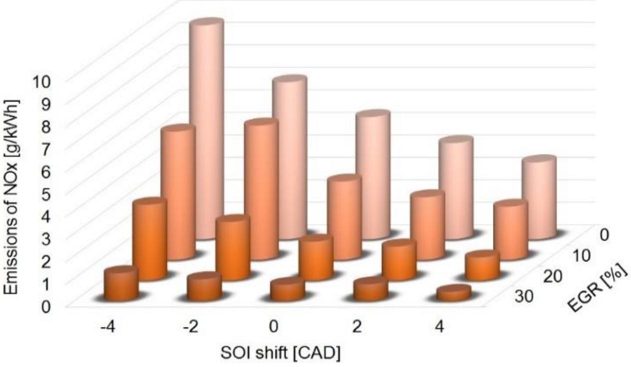

Fig. 7. Indicated specific emissions of NO_x for DF at variable SOI and EGR rates

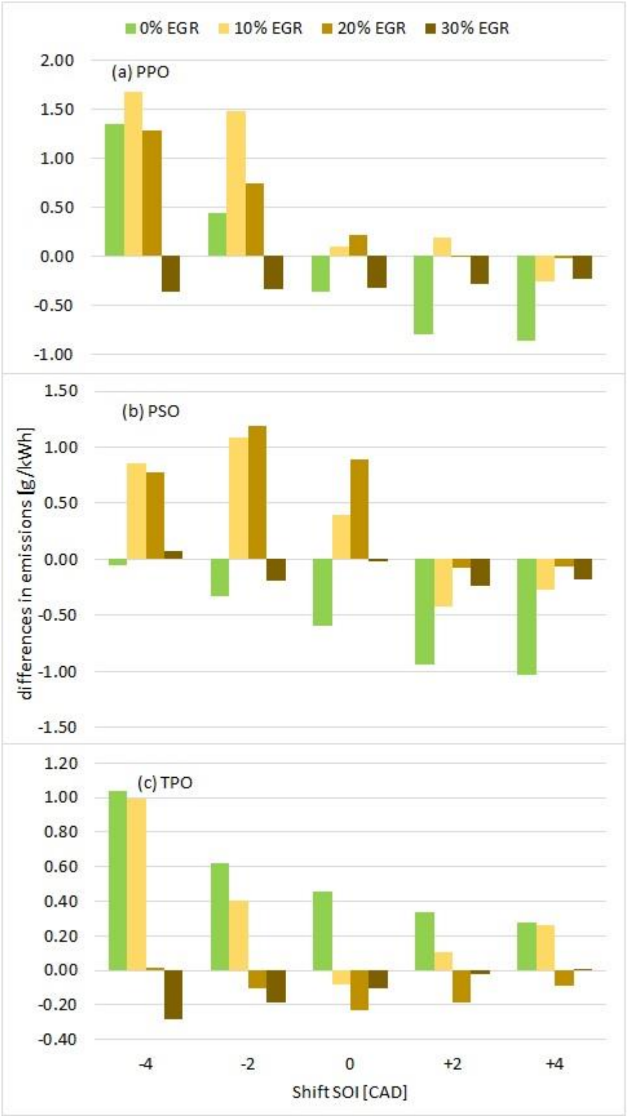

Fig. 8. Differences in NO_x emissions between DF and tested WPO at variable SOI and EGR rates

4. Conclusions

Presented outcomes of thorough comparison of the Common Rail diesel engine operating parameters, were focusing on the use of WPO fuels. The following requests were received:

1. Creating blends of WPO with DF produces fuels that meet stringent fuel standards in all parameters except sulfur content.
2. With split fuel injection, diminished pyrolysis fuels CI is mainly manifested by delayed ignition of the pilot fuel. Introduction of WPO causes a delay in the commencement of combustion by up to 3 CAD.
3. NO_x emissions do not depend on the used fuel blends.
4. The high content of aromatics in PSO causes increased formation of AHC.
5. The high sulfur content in TPO excludes this fuel from direct use in mixtures with DF. Additional desulfurization is necessary after the pyrolysis process.
6. TPO can realize low-temperature combustion, partially pre-mixed, across a broader range of the engine working area. The favorable NO_x/PM level extends to elevated values of EGR and early injection modes due to the prolonged delay of TPO ignition . Its promote the formation of a pre-mixed charge before auto-ignition.
7. PSO can achieve a PM emission reduction of 0.21 g/kWh compared to DF with maintain indicated efficiency target of 42%.
8. The low viscosity of PPO combined with low reactivity causes excessive mixing of the charge in the cylinder before combustion begins. This is manifested by increased CO and HC emissions.

In summary, the obtained results clearly indicate that there are no restrictions on the use of PPO oils in blends with DF. Observed combustion delay can be eliminated by engine calibration. At the same time, limitations were observed in the use of PSO oils due to high aromatic content resulting in high AHC emissions. In the case of TPO oil, its high sulfur content precludes its use as a fuel for road vehicles. However, the path to non-road applications remains open, but requires further research on reducing sulphur compounds.

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Nomenclature

AHC	aromatic hydrocarbon
CI	compression ignition
CO	carbon monoxide
DF	diesel fuel
EGR	exhaust gas recirculation
ELT	end-of-life tires
FAME	fatty acid methyl esters
FTIR	Fourier-transform infrared
HC	hydrocarbon emissions
HRR	heat release rate
HVO	hydrotreated vegetable oil
IMEP	indicated mean effective pressure
MFB	mass fraction burned
NO_x	nitrogen oxides
PM	particulate matter
PP	polypropylene
PPO	polypropylene oil
PS	polystyrene
PSO	polystyrene oil
SO_2	sulfur dioxide
SOI	start of injection
TDC	top dead center
TPO	tire pyrolysis oil
WPO	waste pyrolytic olis

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