Comparative analysis of bio-intermediates and waste-derived fuels in experimental gas turbine

Žiga Rosec[1], Véronique Dias[2], Francesco Contino[2], Tomaž Katrašnik[1], Tine Seljak[1*]

[1]Faculty Of Mechanical Engineering, University Of Ljubljana, Slovenia, [2]Institute of Mechanics, Materials and Civil Engineering, Catholic University of Louvain, Belgium

Submitted to Journal:
Frontiers in Mechanical Engineering

Specialty Section:
Engine and Automotive Engineering

Article type:
Original Research Article

Manuscript ID:
546914

Received on:
30 Mar 2020

Frontiers website link:
www.frontiersin.org

Conflict of interest statement

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest

Author contribution statement

The experiments were designed and conducted by TS. ŽR conducted data post processing and writing. TS provided the overall idea and relevance of the work. Writing was principally performed by ŽR, TS and TK, essential contributions were provided by VD and FC in the discussions of the results. TK, FC and TS provided guidance in reviewing of the paper.

Keywords

Bioliquids, Oxygenated fuels, Emissions, Particulate Matter, gas turbine, Waste-derived fuels, combustion

Abstract

Word count: 342

The paper presents a first comparative analysis of emission formation phenomena of three different bioliquids, derived from low cost waste streams while utilizing the same, gas turbine based experimental setup. A consistent and unbiased comparison is ensured by the application of the same experimental test rig featuring only those minor fuel based adaptions, which are required to ensure most favourable operation of each of the analysed fuels. This provides the direct comparative data between combustion performance of liquefied wood, obtained through solvolysis process, glycerol and waste liquor from nanocellulose production that were previously tested in various combustion systems, hence making direct evaluation of fuel's suitability difficult. The study focuses on analysis of all key thermodynamic parameters, significant emission species covering CO, NOx, HC particulate matter and soot as well as identification of underlying phenomena for observed emission trends. These indicate that for NOx emissions a good correlation exists to stoichiometric ratio of the fuels, where low stoichiometric ratio results in lower NOx emissions, provided oxygen content is the main diluent and fuel bound nitrogen is low. As all tested fuels feature oxygen content above 43%, this enables a large improvement in NOx – CO trade off, as CO emissions are reducing with higher peak combustion temperatures while minimally increasing NOx emissions. Similar observations are made for particulate matter – NOx trade off, however the ash content significantly impacts the particulate matter emission, hence reducing the potential for clean combustion of waste liquor. In the case of glycerol with no ash content, soot emissions are minimal and for an order of magnitude lower than for benchmark diesel fuel, as there are numerous phenomena effectively reducing their formation and increase their oxidation. The presented research confirms that utilization of bio-intermediates and waste-derived fuels in appropriate combustion setups can beside low CO2 footprint, feature also very low emissions of other pollutant species, providing that fuels feature high oxygen content, low ash content and low nitrogen content. With such approach it is possible to achieve clean combustion that is fully in line with circular economy guidelines.

Contribution to the field

The paper presents a first comparative analysis of emission formation phenomena of three different bioliquids, derived from low cost waste streams while utilizing the same, gas turbine based experimental setup. A consistent and unbiased comparison is ensured by the application of the same experimental test rig featuring only those minor fuel based adaptions, which are required to ensure most favourable operation of each of the analysed fuels. This provides the missing link between combustion performance of liquefied wood, obtained through solvolysis process, glycerol and waste liquor from nanocellulose production that were previously tested in various combustion systems, hence making direct evaluation of fuel's suitability difficult. The presented research confirms that utilization of bio-intermediates and waste-derived fuels in appropriate combustion setups can beside low CO2 footprint, feature also very low emissions of other pollutant species, providing that fuels feature high oxygen content, low ash content and low nitrogen content. With such approach it is possible to achieve clean combustion that is fully in line with circular economy guidelines.

Ethics statements

Studies involving animal subjects
Generated Statement: No animal studies are presented in this manuscript.

Studies involving human subjects
Generated Statement: No human studies are presented in this manuscript.

Inclusion of identifiable human data
Generated Statement: No potentially identifiable human images or data is presented in this study.

Data availability statement

Generated Statement: The datasets presented in this article are not readily available because Intellectual property rights prevent the publishing of raw data.. Requests to access the datasets should be directed to tine.seljak@fs.uni-lj.si.

Comparative analysis of bio-intermediates and waste-derived fuels in experimental gas turbine

Tine Seljak[1*], Žiga Rosec[1], Véronique Dias[2], Francesco Contino[2], Tomaž Katrašnik[1]

[1]Laboratory for internal combustion engines and electromobility, Faculty of mechanical engineering, University of Ljubljana, Ljubljana, Slovenia

[2]Institute of Mechanics, Materials and Civil Engineering, Thermodynamics and Fluid mechanics Division, Université catholique de Louvain, Louvain-la-Neuve, Belgium

*** Correspondence:**
Corresponding Author
tine.seljak@fs.uni-lj.si

Keywords: Bioliquids[1], Oxygenated fuels[2], Emissions[3], Particulate matter[4], Gas turbine[5], Waste-derived fuels[6], Combustion[7].

Abstract

The paper presents a first comparative analysis of emission formation phenomena of three different bioliquids, derived from low cost waste streams while utilizing the same, gas turbine based experimental setup. A consistent and unbiased comparison is ensured by the application of the same experimental test rig featuring only those minor fuel based adaptions, which are required to ensure most favourable operation of each of the analysed fuels. This provides the direct comparative data between combustion performance of liquefied wood, obtained through solvolysis process, glycerol and waste liquor from nanocellulose production that were previously tested in various combustion systems, hence making direct evaluation of fuel's suitability difficult. The study focuses on analysis of all key thermodynamic parameters, significant emission species covering CO, NO_x, HC particulate matter and soot as well as identification of underlying phenomena for observed emission trends. These indicate that for NO_x emissions a good correlation exists to stoichiometric ratio of the fuels, where low stoichiometric ratio results in lower NO_x emissions, provided oxygen content is the main diluent and fuel bound nitrogen is low. As all tested fuels feature oxygen content above 43%, this enables a large improvement in NO_x – CO trade off, as CO emissions are reducing with higher peak combustion temperatures while minimally increasing NO_x emissions. Similar observations are made for particulate matter – NO_x trade off, however the ash content significantly impacts the particulate matter emission, hence reducing the potential for clean combustion of waste liquor. In the case of glycerol with no ash content, soot emissions are minimal and for an order of magnitude lower than for benchmark diesel fuel, as there are numerous phenomena effectively reducing their formation and increase their oxidation. The presented research confirms that utilization of bio-intermediates and waste-derived fuels in appropriate combustion setups can beside low CO_2 footprint, feature also very low emissions of other pollutant species, providing that fuels feature high oxygen content, low ash content and low nitrogen content. With such approach it is possible to achieve clean combustion that is fully in line with circular economy guidelines.

1 Introduction

The research area of alternative fuels was previously heavily boosted by the first EU RED directive (*Directive (EU) 2009/28/EC of the European Parliament and of the Council of 23 April 2009 on the Promotion of the Use of Energy from Renewable Sources* 2009), that fostered innovation and implementation efforts in the EU as well as worldwide. Within the first RED perspective, several commercial setups for biofuel production were put in function, however they mostly relied on 1st generation feedstock, where significant market share was obtained with biodiesel (Ciriminna et al., 2014). Along with these, a strive continued to reach commercially feasible setups for production of advanced biofuels with 2nd generation and waste-derived feedstocks. In the first stage of development process they were capable of producing bio-intermediates, a mid-process products that required substantial upgrading before utilization as fuels (Xu et al., 2018). They were defined within RED as bioliquids (liquid fuels made from biomass for energy purposes other than transport). Although these are not suitable for transportation, low price and high maturity of technology makes them a viable alternative for stationary power generation and for cogeneration (heating and cooling).

Within the recast of RED to RED II *(Directive (EU) 2018/2001 of the European Parliament and of the Council of 11 December 2018 on the Promotion of the Use of Energy from Renewable Sources. 2018)*, bioliquids still play an important role as they count towards the goal for achieving sufficient renewable energy sources. Within these, currently technically confirmed bioliquids that are suitable for power generation comprise pyrolysis oils from biomass (Hita et al., 2016), liquefied biomass obtained through solvolysis process (Seljak et al., 2012) and a number of other niche products that originate as side-streams in various production processes with a most notable case being crude glycerol that originates as a side stream of biodiesel production (Mishra and Goswami, 2018). Although being a promising bio-intermediate for further upgrading, hydrothermal liquefaction of biocrude (Dimitriadis and Bezergianni, 2017) was not technically proven to be directly usable in power generation systems, however this does not limit its significant potential for upgrading and refinement to transportation fuels (Tzanetis et al., 2017). The above bioliquids are almost exclusively relying on waste-based feedstock, hence they do not interfere with food-supply chain and do not exhibit indirect land use change risk, what is often the case with straight vegetable oils, which also classify as bioliquids but do not fit very well within circular economy.

The relatively low cost of bioliquids and bio-intermediates (20 €/GJ for pyrolysis oil, 14€/GJ for HTL biocrude and 11€/GJ for crude glycerol) in comparison to 37 €/GJ for ethanol or 19 €/GJ for biodiesel, lead to thorough investigation of power generation possibilities. The research was mostly oriented towards power generation with pyrolysis oils, where small scale gas turbines (Buffi et al., 2018; Beran and Axelsson, n.d.; Sallevelt et al., 2014; Pozarlik et al., 2015; Cappelletti et al., 2013) and reciprocating engines (Chiaramonti et al., 2007) were often researched. Glycerol, for example was also investigated as a fuel for boilers, however promising results were obtained in gas turbines as well (Seljak and Katrašnik, 2019). Liquefied lignocellulosic biomass was investigated in experimental gas turbine systems and small-scale gas turbines exclusively (Seljak et al., 2014; Buffi et al., 2018). All of the studies used a wide variety of experimental setups with different methodologies of experimental evaluation for each of the investigated fuels, hence making a direct comparison of performance difficult. This leads to challenging identification of technical, economical and life-cycle benefits what is of utmost importance when evaluating the environmental suitability of different power generation approaches.

The present study is filling this gap by providing the first thorough comparative analysis of three different bio-intermediates and waste-derived fuels from industrial processes that are strictly following the circular economy guidelines and feature low CO_2 equivalent (according to RED II, crude glycerol from biodiesel production can be considered to have CO_2 equivalent of zero). Their utilization in power generation at the same time increases the economic viability of baseline processes where they are generated. The comparative analysis for the first time provides an unbiased insight into thermodynamic and emission performance within the same experimental setup and provides a long needed link between the currently available studies on separate fuels. The analysed fuels comprise:

- Lignocellulosic biomass, liquefied through solvolysis process that was first proposed as a fuel in (Rezzoug and Capart, 2002) and experimentally confirmed in (Seljak et al., 2012) and uses wood trimmings, residual lignocellulose materials and reclaimed wood as a feedstock.
- Glycerol, used as a model compound for crude glycerol originating as a side stream from biodiesel production, which exhibits market price similar to natural gas (11 €/GJ) and presents a well known burden to biodiesel industry.
- Waste Liquor originating as a side stream from production of nanocrystallinic cellulose that currently has no practical use and is discarded at high costs. Successful implementation of waste liquor as a fuel can lead to the first zero-waste nanocellulose production process.
- Diesel fuel, used as a benchmark to provide direct comparison of performance to above bioliquids.

All investigated fuels feature challenging physical and chemical properties with high viscosity and high oxygen content being the key limiting factors for utilization in a single experimental setup. To overcome this, a modular, gas turbine based system, specifically tailored for low calorific value fuels with high viscosity was developed. It features above state of the art fuel flexibility and enables analysis of all key thermodynamic parameters and most significant emission species including CO, NO_x, HC and particulate matter. On this basis, the study thus provides comparative links of underlying phenomena that is present during combustion and emission formation of different bioliquids which can be extrapolated to gas turbine based systems. Featured analysis is thus providing an insight into environmental performance of investigated bioliquids and is a key enabler for further technical advances of baseline processes that are oriented towards zero-waste and materially independent principles.

2 Fuel processing

The following section provides detailed information on fuel processing in order to exhibit the relative complexity of different approaches and opportunities that are available in the area of initial feedstock used for production of fuels. Further, the section thoroughly describes experimental procedure with emphasis on key required subsystems that enable the unified analysis of fuels with pronounced relative deviation of physical and chemical properties.

2.1 Liquefied wood

The process for obtaining liquefied wood is well developed and optimized in several earlier studies (Jasiukaityte-Grojzdek et al., 2012) . Optimized formulation is produced by preparing a finely ground spruce wood (with particle size approximately 3mm) from trimming residues. Ground wood is then introduced to cold mixture of multifunctional alcohols (glycerol and diethylene glycol in 1:1 ratio), where wood to multifunctional alcohols ratio is 1:3 by mass. 3% of multifunctional alcohols mass is

substituted with para toluensulfonic acid that acts as a catalyst. The liquefaction procedure involves heating of the mixture to 160°C with simultaneous ultrasonic agitation to speed up the lignocellulosic complex break down (Kunaver et al., 2012). After 60 minutes, the mixture is cooled to room temperature in order to prevent formation of recondensation products. Besides depolymerized cellulose, liquefied wood contains also lignin degradation products that result in notable content of cyclic hydrocarbons. Additionally, it is highly viscous and prone to thermal degradation that is a consequence of restarted liquefaction process at elevated temperatures due to catalyst being still present in the final product. The overall energy requirement for production amounts to 2% of fuel's heating value, while liquefaction yield is above 98%. Figure 1 presents the final product, the key properties are presented in Table 1.

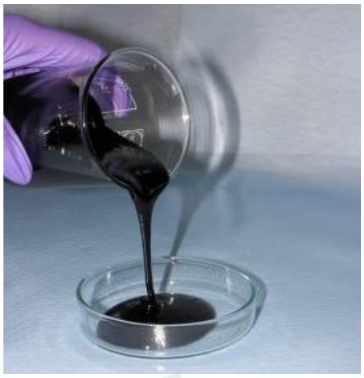

Figure 1: Liquefied wood

2.2 Glycerol

Glycerol is an abundant side-product of the transesterification process that amounts to approximately 10% of the biodiesel produced and is perceived as one of the major bottlenecks due to large quantities produced (Monteiro et al., 2018) and low price (Ciriminna et al., 2014). Depending on the process parameters and feedstock, the properties are highly variable in terms of catalyst residuals and mineral matter content, water content and methanol content (Kumar et al., 2019). Major contaminants are sodium, calcium, potassium, magnesium and phosphorous. Many process variations increase the purity of the co-products, for example fixed bed catalyst. For the sake of brevity of the paper, possible process variations to increase the purity of side-product will not be covered here. Nevertheless, it is worth mentioning that fixed bed catalysts are yielding glycerol with lowest contaminant content. Depending on the final use, crude glycerol is usually refined to different levels of purity. For power generation purposes, the key challenge is the mineral content, whereas water content and methanol content are to moderate extent tolerated, since they aid at reduction of viscosity, which is a key parameter for atomization of the fuel, and aid at ignition properties. To provide unbiased and transferrable comparative analysis with other investigated fuels, purified glycerol was used in the study with properties presented in Table 1.

2.3 Waste Liquor

One of possible processes for production of nanocrystalinic cellulose (NCC) follows the similar route as for production of liquefied wood, however, for optimizing the NCC yield, cotton linters are used as a feedstock and several additional steps are involved for isolation of NCC. These involve dilution with dioxane and centrifuging to sediment the nanocrystallinic particles. After centrifuging, waste liquor is obtained that is recycled through the process several times in place of fresh multifunctional alcohols mixture. The process route is presented in Figure 2 and further details are described by

(Kunaver et al., 2016). The key difference to liquefied wood is the use of cellulose rich feedstock (waste textiles, residual cotton linters,…) that leads to little to no cyclic hydrocarbon content in waste liquor. At the same time, the process is optimized to each feedstock by using tailored multifunctional alcohol mixture, liquefaction time as well as other parameters. As above described glycerol and liquefied wood, waste liquor features high oxygen content as well, however viscosity exhibits less dependence on temperature which is to certain extent limiting the useful fuel preheating temperatures. With utilization of waste liquor for power generation, a possibility to set up a first zero waste process for nanocellulose production emerges, which is a key enabler for large scale nanocellulose production, since all of the processes produce large quantities of side-streams with no further use (Brinchi et al. 2013). Key properties of waste liquor are presented in Table 1.

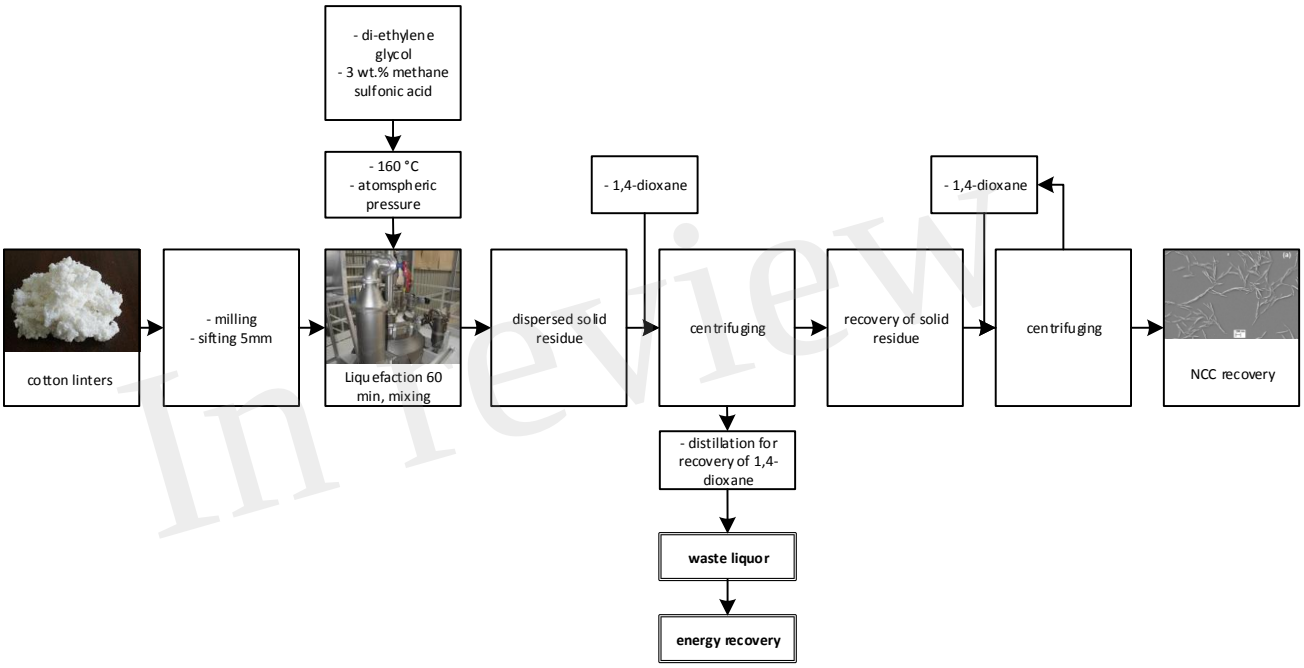

Figure 2: Waste liquor origin

Table 1: Properties of investigated fuels

	Glycerol	Liquefied wood	Waste liquor	Diesel
C (wt%)	42.19	47.52-47.6	44.3	87.00
H (wt%)	9.14	7.98-8.00	9.0	13.00
N (wt%)	0	0.19-0.34	0	0
S (wt%)	0	0.89	0.9	<0.001
O (wt%)	48.67	43.26-43.34	45.9	0
Ash content (wt%)	0.0002	0.1	0.6	/
Density (kg/L)	1.26 at 20°C	1.3 at 15°C	1.25 at 15°C	0.82-0.845 at 15°C
LHV (MJ/kg)	19	20.2	22.6	42,2
Stoich. Ratio	5.19	6.8	6.19	14,7

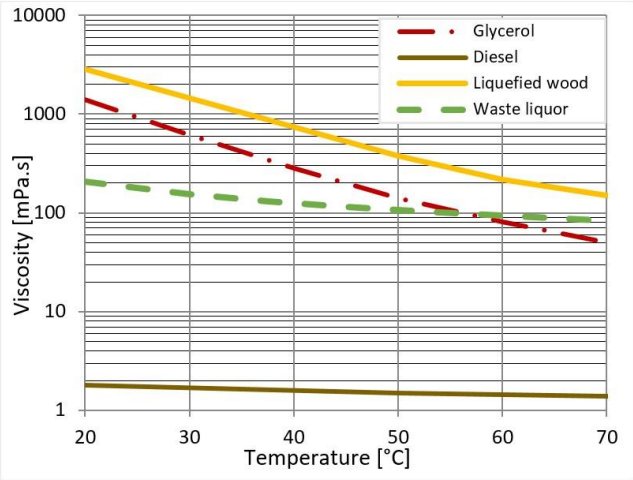

Figure 3: Viscosity dependence on temperature for the investigated fuels.

All presented fuels that will be investigated through this study exhibit similar challenging chemical and physical properties, although they are not interrelated. One of the challenging properties is the high viscosity presented in Figure 3 in comparison to a standard diesel fuel, which limits the atomization performance of the fuels and hence negatively impact the mixture formation process. Furthermore, their viscosities are temperature dependant, suggesting the temperature can be exploited to reduce the viscosity. Even though they are derived from different sources they all stem from waste-streams, which presents a unique possibility for renewable energy generation while at the same time reducing waste streams and enabling zero waste production processes.

3 Experimental setup for combustion analyses

3.1 Combustion test rig

Evaluating the combustion capabilities of the investigated fuels is done on the experimental rig presented in Figure 4. The experimental rig can be easily modified to accommodate the changes in fuel type used, a detailed description is given by (Seljak and Katrašnik, 2016).

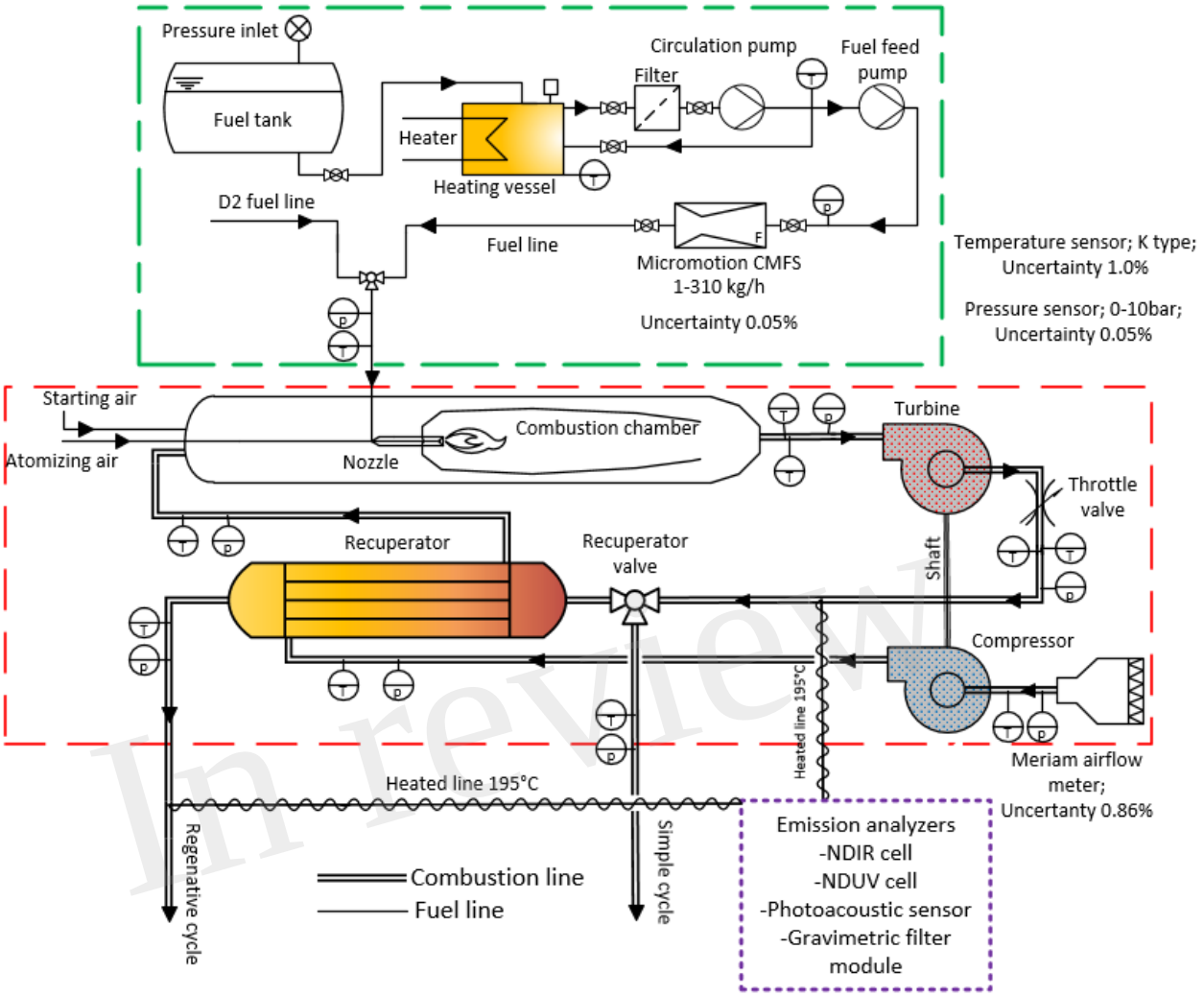

Figure 4: Scheme of the experimental rig used for evaluating combustion performance of evaluated fuels.

Fuel preparation subsystem is marked within the green rectangle, where different fuels are connected from their individual fuel tank to the heating vessel. Downstream, the circulation pump establishes a homogenous fuel temperature and heats up the fuel line with possible inclusion of a fuel filter to remove possible impurities. The heated fuel is then fed to the combustion chamber with the fuel feed pump through a coriolis mass flow meter. The fuel processing subsystem is fully pressurized to prevent the boil-off of volatile components while at the same time features low heater surface temperatures in order to avoid thermal degradation and phase separation of the fuels, which are due to a large content of oxygenated groups thermally unstable.

The combustion test rig (within the red rectangle) is the experimental subsystem that is designed for combustion analyses, hence the power output is not measured. Different power outputs can be emulated either via throttle valve that reduces the enthalpy difference on the turbine or via fuel mass flow control. Both approaches result in altered turbine inlet temperature (TIT), a characteristic parameter for defining operating point of the experimental system. The main components also include a single can combustion chamber and the recuperator which enables regenerative cycle operation and hence primary air temperatures in excess of 450°C. The starting procedure uses diesel fuel. Ignition is achieved with a pilot diesel flame in simple cycle mode, which is followed by a

gradual increase in regeneration intensity to ultimately achieve operation in regenerative cycle. After operating temperatures are stabilized the switch from diesel to investigated fuel is performed. From this point, the experimental systems operates with the investigated fuel, including all transient operation and stabilization intervals.

3.2 Measurement equipment and post processing

Seven type-K thermocouples, 5 low-frequency piezoresistive pressure sensors and laminar flow airflow meter are positioned on all characteristic points of the experimental rig. Positions are discernible from Figure 4. In the emission analysers NO_x emissions are measured in a NDUV cell and CO, CO_2 and HC in a NDIR cell.

The values for each individual operation point is averaged form 30 seconds of recorded data in stabilized operational point. All the evaluated emissions are normalized to fuel power, to assure a credible comparison. The mathematical formulation for any normalized emission is defined as:

$$\boldsymbol{E_{x\,normalized} = \left(\frac{E_x \cdot M_x}{(1000000 - M_x) \cdot M_{exhaust\,gas}}\right) \cdot (\dot{m}_{fuel} + \dot{m}_{air}) \cdot \left(\frac{1000}{LHV/_{3,6} \cdot \dot{m}_{fuel}}\right)} \quad (1)$$

$E_{x\,normalized}$ represents the normalized emissions while E_x represents the measured values of the chosen emission, M_x is the corresponding molar mass and $M_{exhaust\,gas}$ the molar mass of exhaust gases. Mass flows of fuel and air are represented as $\dot{m}_{fuel}$ and $\dot{m}_{air}$ and LHV the lower heating value of fuel.

The analysis and evaluation of data follows the emission concentrations normalized to fuel power at each distinctive operational point with different fuels. The environmental impact is assessed through CO, NOx, HC (as C_6H_{12}) and particulate matter results. While direct footprint of combustion can be estimated via combined concentrations, the Results and discussion section specifically discusses each emissions species separately in order to provide an insight into combustion phenomena that is responsible for observed trends and to establish a link between analysed fuels and emission performance.

Particulate matter (PM) emissions are measured with a PM-PEMS analyser produced by AVL List GmbH. It is capable of accurately determining the PM emissions using two subsystems (photoacoustic sensor and gravimetric filter module). The exhaust sample is firstly diluted to a ratio between 2 and 50 with filtered air. The photoacoustic sensor is capable of determining the soot concentration using the photoacoustic method where strongly absorbent soot particles are exposed to a modulated laser beam. This induces periodic expansion and contraction as the particles get warmed and cooled, what results in sound wave generation which is measured with a microphone determining soot concentration. Calculation of cumulative soot mass on filter is done from the continuous soot signal and filter flow from start to end of filter loading presented in equation (2). This parameter is then used to calculate the scaling factor (fac) with the filter mass in equation (3).

$$\boldsymbol{m_{soot}(filter) = \int_0^{t_{fil}} \left(c_{soot}(t) \cdot \left(\frac{q_{fil}(t)}{60 \cdot (1000)}\right)\right) dt} \quad (2)$$

m_{soot}(filter) represents the cumulative soot mass on filter, t_{fil} is the time the filter is loaded, c_{soot} is the continuous soot signal and q_{fil} is the filter flow.

$$fac = \frac{m_{fil}}{m_{soot}(filter)} \quad (3)$$

In equation (3) fac represents the scaling factor, m_{fil} is the filter mass flow and the m_{soot}(filter) presents the previously calculated cumulative soot mass on filter.

$$c_{PM}(t) = c_{soot}(t) \cdot fac \cdot r_d(t) \quad (4)$$

To calculate the real-time PM emissions with the scaling factor, we use equation (4), where the dilution ratio (r_d) is taken into account.

4 Results and discussion

The following section presents the results, where emphasis is given on emission data. Results on thermodynamic parameters are provided to ensure unbiased comparative analysis and evaluation of possible impact on thermodynamic performance. As all experiments were carried out on the same experimental rig, the differences in emission concentrations can mainly be attributed to fuel physical and chemical properties and only to minor extent to minimal changes that were implemented in experimental setup to accommodate the use of different fuels. Emission concentrations are plotted versus TIT at different fuel temperatures.

4.1 Comparative analysis of thermodynamic parameters

The baseline thermodynamic parameters, pressure ratio and primary air temperature, need to be evaluated to assure a comparative analysis. TIT is the leading parameter for establishing operational points that is controlled with fuel flow that results in higher pressure ratios. In general, the established conditions mirror those present in commercial micro turbines. In Figure 5 the pressure ratios are presented for each separate fuel. The change in fuel temperature does not affect the pressure ratios, also the relative differences between investigated fuels are small what is a result of minor changes is in the experimental set up. A slightly larger difference is observed with diesel fuel. As the experimental rig was purposely build for testing highly viscous fuels with low LHV, benchmark data with diesel fuel was obtained in a separate set of experiments with partially closed throttle valve position after the turbine, hence reducing the pressure ratio slightly. Simultaneously, the fuel flow was reduced to maintain comparable TIT and PAT as with other investigated fuels, however the emission concentrations are normalized to fuel power, hence eliminating this effect.

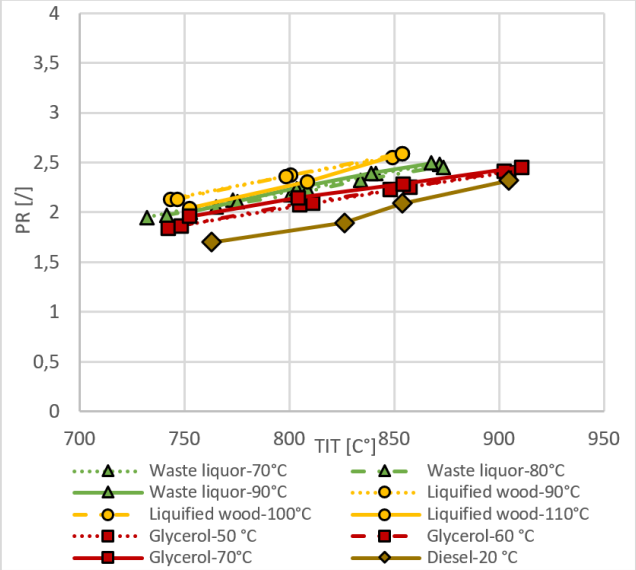

Figure 5: Pressure ratios

With increasing TIT, impact on primary air temperature is visible as well. This happens as higher TIT results in higher turbine outlet temperature and hence higher temperature of regenerated primary air. The primary air temperature shown in Figure 6, does not change significantly for each individual fuel, with minorly lower primary air temperature obtained with liquefied wood which is a consequence of slightly higher pressure ratio obtained with liquefied wood in Figure 5. The reason for this is in minor changes in experimental setup that provided lower pressure drops on the intake side when utilizing liquefied wood. Diesel fuel shows comparable primary air temperatures to other fuels although the pressure ratio is slightly lower. The reason for this can be traced back to reduced air mass flow that impacts the primary heat exchanger effectiveness. The detailed thermodynamic analysis for this phenomena is not discussed here as the emissions of diesel fuel are given for a reference purpose and all emission concentrations are normalized to fuel power.

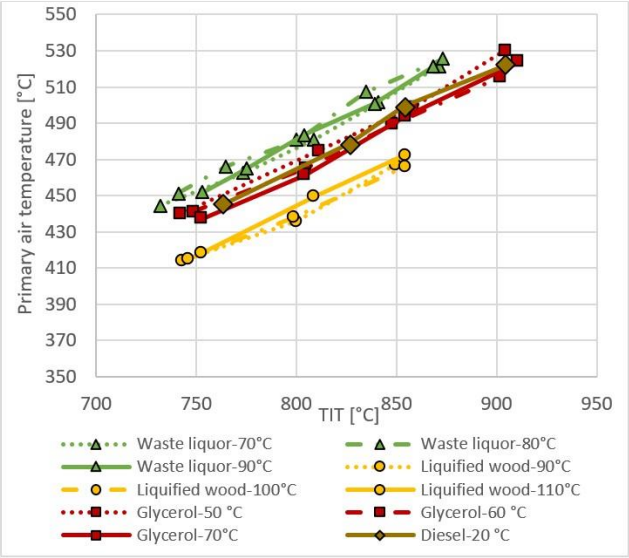

Figure 6: Primary air temperatures

4.2 NO_x emissions

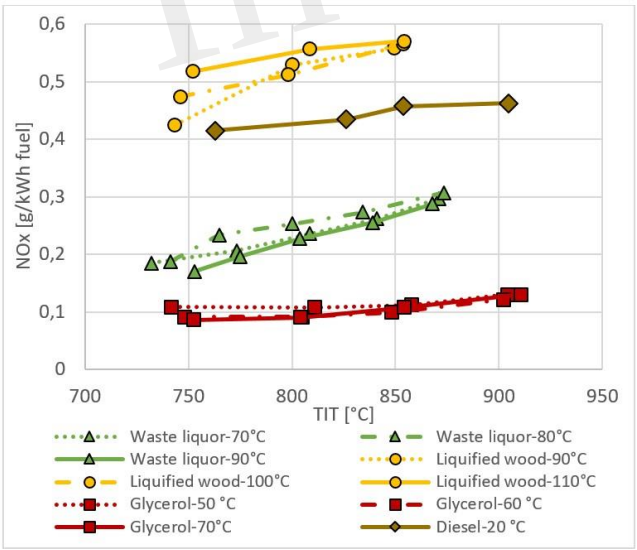

Figure 7: NO_x emissions for different fuels at different fuel temperature

NO_x emissions for each investigated fuel are shown in Figure 7. None achieve similar emissions, but all exhibit increased NO_x emissions with increasing TIT. In addition, the fuel temperatures play little to no role in NO_x emissions formation as no trends are immediately observed, apart from liquefied wood, which features highest viscosity and at low TIT and low fuel preheating temperatures exhibits higher NO_x emissions what is a consequence of large fuel droplets that feature isolated combustion, hence providing large cumulative flame front area, where NO_x formation is promoted.

The investigated fuels are all highly oxygenated and have similar characteristics, surprisingly their effect of inhibiting the mechanisms responsible for NO_x varies from fuel to fuel. The high oxygen content is proven to be beneficial in regard of emission reduction (Nabi 2010), as glycerol and waste liquor fuels emissions are 2-fold lower for waste liquor and 4-fold lower for glycerol compared to the reference diesel fuel. Again, liquefied wood exhibits higher NO_x emissions what can be linked to high nitrogen content in the fuel.

The high oxygen content in fuel is linked to low stoichiometric ratios, which is the main mechanism responsible for lower NO_x emissions. If low stoichiometric ratio is a consequence of high oxygen content, what is the case in investigated fuels, smaller quantities of air are required to achieve flammability limits and consequently less nitrogen is introduced into the high temperature zones to participate in the thermal (Heywood, 1988) and also prompt (Fenimore, 1971) mechanisms responsible for NO_x emissions. This is the main advantage of glycerol for achieving the lowest NO_x emissions, due to having the lowest stoichiometric ratio followed by waste liquor.

The fuel bound oxygen can also affect the adiabatic flame temperature (Nabi, 2010). Increasing the oxygen content will decrease the adiabatic flame temperature, resulting in decreased hot zones and reduced thermal formation of NO_x.

Even though the beneficial effects of fuel bound oxygen are present with liquefied wood, the NO_x emissions exceed those of diesel, what can be linked to another mechanism of NO_x formation, as liquefied wood is the only investigated fuel that contains traceable amount of nitrogen. This fuel bound nitrogen forms NO_x emissions (Buffi et al., 2018), which results in inferior performance of liquefied wood in comparison to the other fuels that have no nitrogen content. We can assume that the liquefied wood would still produce higher NO_x emissions even if no fuel bound nitrogen would be present, as it has the highest stoichiometric ratio of all investigated fuels.

4.3 CO emissions

Considering the high viscosity and density of investigated fuels, the observed CO emission trends are to some extent expected. Particularly viscosity and density notably impact the mixture formation process, resulting in increased CO emissions. The main reason is reduced atomization performance and larger droplets are formed that feature lower surface to volume ratio which is additionally influenced by high density of the fuel and causes even lower mass to volume ratio. This, combined with large droplet momentum inhibits the residence time of droplets in high temperature zones of combustion chamber and reduces the heat transfer, required for fuel vaporization. With combustion taking place later in the primary zone, a significant amount of reaction quenching can occur that stops the completion of CO to CO_2 reactions.

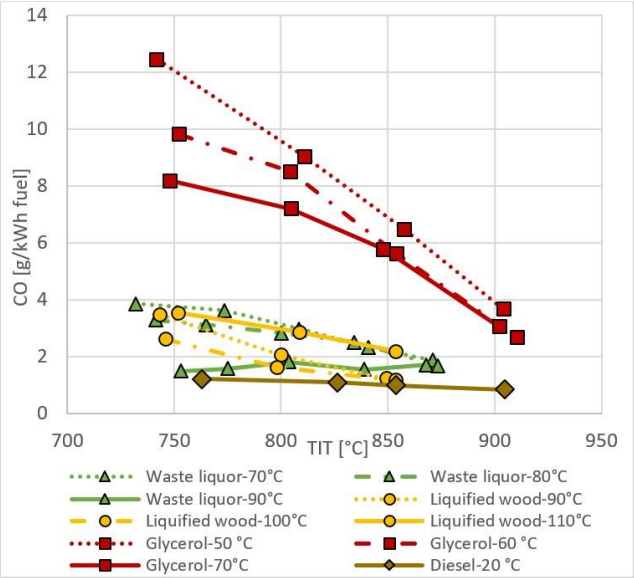

Figure 8: CO emissions for different fuels at different fuel temperatures

In Figure 8, the liquefied wood and waste liquor emissions are in the same range, even though the liquefied wood viscosity is higher and the increased temperature of the fuel brings liquefied wood and waste liquor to similar values resulting in similar CO emissions. Increasing the fuel temperature does have a limit as is observed with liquefied wood as a high fuel temperature of 110°C can reach the fuel nozzle coking point which impairs the atomization ability of the nozzle and increase the CO emissions (Albert-Green and Thomson, 2018).

The large gap between the glycerol and other fuels is not a result of viscosity as glycerol viscosity values sits between those of liquefied wood and waste liquor and even lower at higher temperatures. The higher CO emissions are believed to be connected to the components in fuels. Glycerol is a single-component whereas the liquefied wood and waste liquor are multi-component fuels with large number of different hydrocarbons – from highly volatile to relatively heavy ones with high ignition resistance. Single component composition combined with high boiling point (290°C) and auto-ignition temperature (370°C) of glycerol, drastically alters the evaporation curve. Thus, high temperatures are required to start the vaporization process which increases the delay of mixture formation. Comparing this to the evaporation curves of liquefied wood and waste liquor, where component with lower boiling points help to release the heat early in the mixture formation process and support the vaporization and combustion of other components. This can also explain why increasing TIT has the most effect on lowering the CO emissions with glycerol and is less pronounced with liquefied wood and waste liquor.

The observed trends ultimately suggest that a persistent CO-NO_x trade-off is present among investigated fuels, which is not linked to thermodynamic parameters as they were comparable during the experiments with different fuels. However, as glycerol shows the best NO_x performance and well pronounced reducing trend of CO emission with high TIT, state of the art gas turbine setups that utilize TIT above 1000°C could majorly benefit from the use of glycerol.

4.4 HC emissions

The same problem linked to glycerol, due to being a single-component fuel can also be observed with the HC emissions in Figure 9. Although the spread is smaller, the droplets that do not reach required

temperature conditions are unable to vaporize and fully oxidize. This is also apparent with lower fuel temperatures where the droplets size increases for all fuels, resulting in more HC emissions. Like with CO, a trend of lowering HC emissions is observed for all fuels when increasing TIT. With higher TIT temperatures in the combustion chamber increase, allowing for more favourable conditions for fuel vaporization and combustion reactions to occur. Considering relatively low HC emissions it can be concluded that no significant deviation from baseline diesel fuel is present and HC emissions are not a limiting factor when power generation with investigated waste-derived bioliquids is in question.

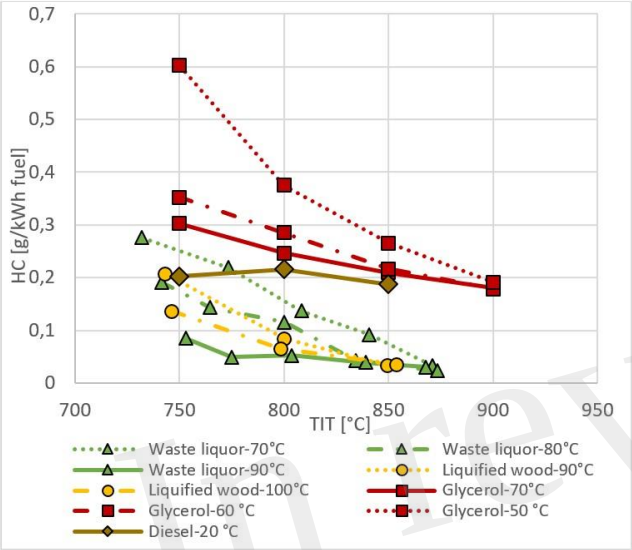

Figure 9: HC emissions for different fuels at different fuel temperatures

4.5 Particulate matter emissions

Particulate matter emissions are measured with two different but correlated methods. As described in section 3, soot emissions are measured with photoacoustic method that is responding to soot particles and is calibrated for diesel fuel. In parallel, the same sample is measured also gravimetrically, which gives the cumulative mass of particulate matter sampled. The real-time concentration of soot is given by photoacoustic method, which is after finalized experiments weighted with data from gravimetric method to obtain real-time particulate matter concentration. This step is done in post-processing.

Ideally, both methods should return similar concentrations. This is the case with baseline diesel fuel, where scaling factor between soot and particulate matter emission is 1.2 since the method is fitted to morphology, optical properties and chemical composition of diesel soot. With other fuels, soot might exhibit significantly different properties that cause lower response to photoacoustic method, particularly due to lower absorbance of irradiated heat and smaller volume change as a consequence of different density and soot composition.

To provide an insight into both measured parameters, soot emission is given in Figure 10 and weighted PM emission is given in Figure 11. Here, no data for liquefied wood is available. However, based on liquefied wood properties, soot formation for liquefied wood might be increased in comparison to waste liquor, as it contains notable amount of cyclic hydrocarbons that act as precursors for soot and originate from lignin degradation products (Jasiukaitytė et al., 2010).

Soot emissions for waste liquor and glycerol is remarkably lower compared to diesel fuel (Figure 10). The diesel soot emissions are increasing with higher TIT as the soot of waste liquor and glycerol decrease. In absolute terms, glycerol exhibits an order of magnitude lower soot emissions than diesel fuel and at least 5-fold lower emissions than waste liquor. Here, fuel bound oxygen plays a vital role as it influences the following phenomena:

1 Altered soot zone formation limits
2 Inhibited formation of soot precursors
3 Increased soot reactivity

The first phenomena responsible for low soot emission trends with glycerol can be linked to high oxygen content in glycerol and purely physical phenomena involved in spray formation. Bonded oxygen becomes during dissociation of glycerol readily available in the flame zone in the form of free radicals, resulting in high local C/O ratio already without any entrapment of external air. Thus in case of GLY the C/O ratio is always below 1 (based on elemental composition of glycerol), regardless of the mixture preparation dynamics, whereas for D2 this value is easily exceeded on the rich side of the mixture, where insufficient air is entrapped into the spray and approaches infinity where only fuel vapour is present. Areas outside the soot formation interval are more likely to occur with glycerol as volume of the mixture within the soot formation limit (EQR between 1,8 and 2,9) is smaller. Similar reasoning is valid also for waste liquor.

The second phenomenon concerns mainly combustion kinetics. Several previous studies have shown that the addition of an oxygen compound to hydrocarbon flames reduces the formation of species considered as "soot precursors", such as acetylene (C2H2), cyclopentadiene (C5H6), benzene (C6H6) and naphthalene (C10H8). These last molecules lead directly to the production of soot. By blending oxygenated compounds into the rich hydrocarbon flames and by keeping the equivalence ratio constant, we observe that hydrocarbon production is governed mainly by the C/O ratio instead of the equivalence ratios. This leads to a lower formation of soot precursors and an increase in the production of light oxygenates (Dias and Vandooren, 2011; Dias et al., 2014). These observations are justified and validated by detailed kinetic analyses that underline the reactivity of reactions with O and OH radicals. In this study, the low C/O ratio of glycerol justifies thus the small amount of soot formation (Figure 10). Moreover, for molecules as complex as glycerol, the multiple carbon-oxygen bonds decrease the formation of soot composed entirely of carbon-carbon bonds. By comparison, diesel is composed only of hydrocarbons with carbon-carbon bonds (alkanes, cycloalkanes, alkenes, aromatic hydrocarbons) which are responsible for the increase in soot (Figure 10). Soot formation is also influenced with temperature, as Figure 10 shows an increase in soot formation for diesel fuel as a function of temperature. Indeed, the production of the first soot particles is promoted by kinetic mechanisms that are very sensitive to temperature. For glycerol and waste liquor, the temperature leads to a more complete combustion which allows a higher production of oxygenated molecules instead of soot emissions.

The third phenomena touches upon the increased reactivity of soot particles, as the oxygenated groups in the investigated fuels have a proven effect on increasing the soot reactivity (Lapuerta et al., 2019), which consequently leads to lower soot in exhaust gases provided long enough residence time is available. This is one of the effects that can be used with gas turbines as the residence time of mixture is longer in comparison to diesel engines. Increased oxidation points become available on the soot surface because of the fuel bound oxygen together with a more porous structure that is more accessible to oxygen attack (Verma et al., 2020), leading to eventual soot burn off. The most important role is played by combustion chamber secondary zone, where gradual reduction of

temperature occurs with high excess air ratios. Under this assumption it is mandatory that secondary air temperature is sufficiently high (in presented case approximately equal as primary air temperature).

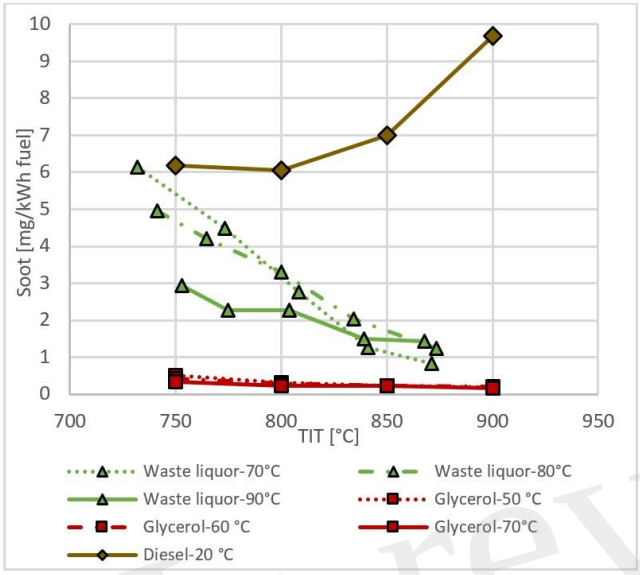

Figure 10: Soot emissions for different fuels at different fuel temperatures

Particulate matter emissions follow similar trends as those of soot emissions with a significant difference in absolute values. The aforementioned scaling factor is 1.2 for diesel fuel, 5.0 for glycerol and 26.0 for waste liquor. This indicates that soot originating from waste liquor combustion exhibits significantly different properties with possible inclusions of matter with low light absorbance, hence gravimetric method detects bigger emitted concentrations than photoacoustic method. This might be linked to significant inclusions of ash in emitted particulate matter, since waste liquor contains 0.6 % of ash. Additional reasons are linked to different soot structure and hence lower response in terms of volume change when particles are heated, hence limiting the sensitivity of photoacoustic method. For glycerol, the scaling factor is much lower what can be linked to the absence of ash.

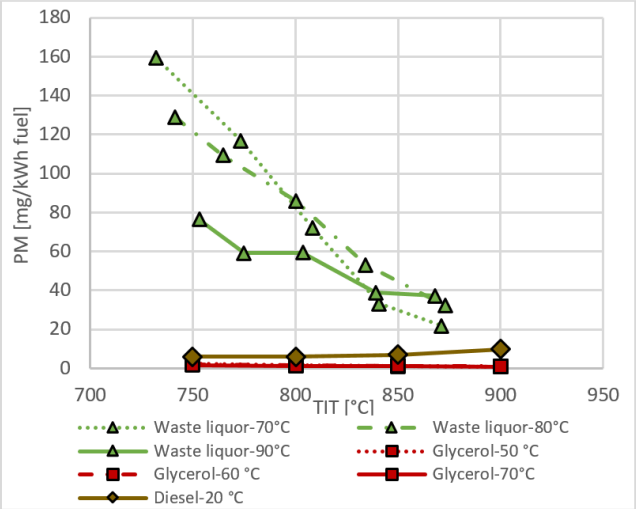

Figure 11: Particulate matter emissions for different fuels at different fuel temperatures

Trend observed with the investigated fuels show that the increase in EQR does not increase the soot or PM emissions, instead the emission trend is reducing as the TIT increases. This offers a promising scenario where with additional increase in TIT that is easily attainable in commercial gas turbine setups, high effective efficiency can be achieved with no or minimal impact on increase in PM emissions. Already the aforementioned improvement in CO – NO_x trade off, similar can also be confirmed with PM – NO_x trade off, making a power generation with low cost, waste-derived bioliquids a promising alternative, providing that ash content is minimized. This puts a new perspective on the waste derived fuels, as they are generally depicted as troublesome fuels with increased emissions and opens a great opportunity for reducing or even closing a fraction of waste streams that originate in different production processes.

5 Conclusions

The study represents the first comparative analysis of different bio-intermediates and waste-derived fuels that originate from established processes in wood industry, biodiesel production and nanocellulose production. All tested fuels originate from renewable sources and their utilization closely follows circular economy guidelines. The result show that improvement of emission footprint of such fuels is not solely the consequence of low CO_2 footprint, but also significant reduction of major pollutant species, namely NO_x and particulate matter.

The NO_x emissions reduction is one of the main advantages in the case of investigated fuels as both waste liquor and glycerol display far lower NO_x emissions in comparison to diesel fuel with the lowest emissions attained from glycerol as it has the highest oxygen content of all the investigated fuels. The reducing trend is consistent with reductions in stoichiometric ratio of the fuels, providing that fuel bound nitrogen is low. The latter case is observed with liquefied wood, which exhibits slightly higher NO_x emissions in comparison to diesel fuel, while glycerol and waste liquor feature approximately 4-fold and 2-fold reduction in NO_x emissions.

CO emissions of all investigated fuels are higher than for diesel fuel, however the emission are reducing towards higher TIT, suggesting that utilization in modern gas turbine based systems with TIT in excess of 1000°C would provide significantly lower concentrations of CO. The reducing trend can be attributed to very high viscosity of the tested fuels that causes generation of larger droplets

during spray formation, which require higher temperatures for further evaporation and mixture formation. Similar tendencies are present also with HC emissions. Combining this observation with reduced NO_x emissions in comparison to diesel fuel, a significantly improved CO-NO_x trade off can be achieved with glycerol and waste liquor, while liquefied wood exhibits very similar trade off behaviour as diesel fuel at highest tested TIT.

Considerably low PM emissions are achieved with the use of glycerol, as the fuel bound oxygen can influence several phenomena resulting in less soot emissions. The underlying reason is that the C/O ratio is heavily influencing the soot zone formation by supplying sufficient oxygen concentrations already without air entrapment in the spray. Kinetics of soot formation are influenced as well, namely through reduction of soot precursor formation. Furthermore, reactivity of soot is increased, due to porous soot particles with larger active area which enables soot burn off in the secondary zone of combustion chamber. Although the same phenomena is present with waste liquor, the PM emissions far exceed those of glycerol and diesel, which can be linked to high ash content in waste liquor.

The performed comparative analysis confirms that with utilization of waste-derived fuels and bio-intermediates for power generation, CO_2 emissions as well as pollutant emissions can be reduced at the same time. In the investigated fuels, where recycling and reuse routes are highly challenging this can lead to attractive business cases and a step towards zero-waste production processes that are fully in line with circular economy guidelines.

6 Conflict of Interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

7 Author Contributions

The experiments were designed and conducted by TS. ŽR conducted data post processing and writing. TS provided the overall idea and relevance of the work. Writing was principally performed by ŽR, TS and TK, essential contributions were provided by VD and FC in the discussions of the results. TK, FC and TS provided guidance in reviewing of the paper.

8 Funding

The authors acknowledge the financial support from the Slovenian Research Agency (research core funding No. P2-0401 – Energy engineering, project funding Z2-1862 – Zero footprint combustion for green energy generation, and young researcher program funding No. 802-7/2019).

9 Data Availability Statement

The datasets presented in this article are not readily available because intellectual property rights under which they were obtained, prevent the publishing of raw data. Requests to access the datasets should be directed to tine.seljak@fs.uni-lj.si

10 References

Albert-Green, S., and Murray, J.T. (2018). The Effects of Nozzle Design on the Combustion of

524 Wood-Derived Fast Pyrolysis Oil. *Biomass and Bioenergy* 117: 102–114.
525 doi:10.1016/j.biombioe.2018.07.002.

526 Beran, M., and Lars-Uno, A. The Use of Pyrolysis Oil for Gas Turbine Produced Power and Heat.

527 Brinchi, L., Cotana, F., Fortunati, E., and Kenny, J. M. (2013). Production of Nanocrystalline
528 Cellulose from Lignocellulosic Biomass: Technology and Applications. *Carbohydrate Polymers*
529 94, 154–169. doi:10.1016/j.carbpol.2013.01.033.

530 Buffi, M., Cappelletti, A., Rizzo, A. M., Martelli, F., and Chiaramonti, D. (2018). Combustion of
531 Fast Pyrolysis Bio-Oil and Blends in a Micro Gas Turbine. *Biomass and Bioenergy* 115, 174–
532 85. doi:10.1016/J.BIOMBIOE.2018.04.020.

533 Buffi, M., Seljak, T., Cappelletti, A., Bettucci, L., Valera-Medina, A., Katrašnik, T., and
534 Chiaramonti, D. (2018). Performance and Emissions of Liquefied Wood as Fuel for a Small
535 Scale Gas Turbine. *Applied Energy* 230, 1193–1204. doi:10.1016/j.apenergy.2018.08.126.

536 Cappelletti, A., Rizzo, A.M., Chiaramonti, D., and Martelli, F. (2013). CFD Redesign of Micro Gas
537 Turbine Combustor for Bio-Fuels Fueling. In *XXI International Symposium on Air Breathing*
538 *Engines (ISABE)*, 1199–1206. Busan, Korea. doi:10.13140/2.1.4096.1601.

539 Chiaramonti, D., Oasmaa, A., and Solantausta, Y. (2007). Power Generation Using Fast Pyrolysis
540 Liquids from Biomass. *Renewable and Sustainable Energy Reviews* 11, 1056-1086.
541 doi:10.1016/j.rser.2005.07.008.

542 Rosaria, C., Pina, C. D., Rossi, M., and Pagliaro, M. (2014). Understanding the Glycerol Market.
543 *European Journal of Lipid Science and Technology* 116, 1432–39. doi:10.1002/ejlt.201400229.

544 Dias, V., Katshiatshia, H. M., and Jeanmart, H. (2014). The Influence of Ethanol Addition on a Rich
545 Premixed Benzene Flame at Low Pressure. *Combustion and Flame* 161, 2297–2304.
546 doi:10.1016/j.combustflame.2014.03.005.

547 Dias, V., and Vandooren, J. (2011). Experimental and Modeling Studies of C2H4/O2/Ar,
548 C2H4/Methylal/O2/Ar and C2H4/Ethylal/O2/Ar Rich Flames and the Effect of Oxygenated
549 Additives. *Combustion and Flame* 158, 848–59. doi:10.1016/j.combustflame.2011.01.015.

550 Dimitriadis, A., and Bezergianni, S. (2017). Hydrothermal Liquefaction of Various Biomass and
551 Waste Feedstocks for Biocrude Production: A State of the Art Review. *Renewable and*
552 *Sustainable Energy Reviews* 68, 113–25. doi:10.1016/j.rser.2016.09.120.

553 Directive (EU) 2009/28/EC of the European Parliament and of the Council of 23 April 2009 on the
554 Promotion of the Use of Energy from Renewable Sources. (2009). https://eur-
555 lex.europa.eu/legal-content/EN/ALL/?uri=CELEX%3A32009L0028 [Accessed March 4, 2020]

556 Directive (EU) 2018/2001 of the European Parliament and of the Council of 11 December 2018 on
557 the Promotion of the Use of Energy from Renewable Sources. (2018). https://eur-
558 lex.europa.eu/legal-content/EN/TXT/?uri=uriserv:OJ.L_.2018.328.01.0082.01.ENG [Accessed
559 March 3, 2020]

560 Fenimore, C. P. (1971). Formation of Nitric Oxide in Premixed Hydrocarbon Flames. *Symposium*

(International) on Combustion 13, 373–80. doi:10.1016/S0082-0784(71)80040-1.

Heywood, J. B. (1988). Engine Types and Their Operation. *Internal Combustion Engine Fundamentals*. McGraw-Hill College.

Hita, I., Arabiourrutia, M., Olaza,r M., Bilbao, J., Arandes, J. M., and Sanchez, P. C. (2016). Opportunities and Barriers for Producing High Quality Fuels from the Pyrolysis of Scrap Tires. *Renewable and Sustainable Energy Reviews* 56, 745–59. doi:10.1016/j.rser.2015.11.081.

Jasiukaityte-Grojzdek, E., Kunaver, M., and Poljanšek, I. (2012). Influence of Cellulose Polymerization Degree and Crystallinity on Kinetics of Cellulose Degradation. *BioResources* 7, 3008–3027. doi:10.15376/biores.7.3.3008-3027

Jasiukaitytė, E., Kunaver, M., and Crestini, C. (2010). Lignin Behaviour during Wood Liquefaction—Characterization by Quantitative 31P, 13C NMR and Size-Exclusion Chromatography. *Catalysis Today* 156, 23–30. doi:10.1016/J.CATTOD.2010.02.001.

Kumar, L. R., Yellapu, S. K., Tyagi, R.D., and Zhang, X. (2019). A Review on Variation in Crude Glycerol Composition, Bio-Valorization of Crude and Purified Glycerol as Carbon Source for Lipid Production. *Bioresource Technology* 293, 122-155. doi:10.1016/J.BIORTECH.2019.122155.

Kunaver, M., Anžlovar, A., and Žagar, E. (2016). The Fast and Effective Isolation of Nanocellulose from Selected Cellulosic Feedstocks. *Carbohydrate Polymers* 148, 251–58. doi:10.1016/j.carbpol.2016.04.076.

Kunaver, M., Jasiukaitytė, E., and Čuk, N. (2012). Ultrasonically Assisted Liquefaction of Lignocellulosic Materials. *Bioresource Technology* 103, 360–66. doi:10.1016/J.BIORTECH.2011.09.051.

Lapuerta, M., Sánchez-Valdepenas, J., Barba, J., Fernández-Rodríguez, D., Andrés, J. P., and García, T. (2019). Analysis of Soot from the Use of Butanol Blends in a Euro 6 Diesel Engine. *Energy and Fuels* 33, 2265–77. doi:10.1021/acs.energyfuels.8b04083.

Mishra, V. K., and Goswami, R. (2018). A Review of Production, Properties and Advantages of Biodiesel. *Biofuels* 9, 273–289. doi:10.1080/17597269.2017.1336350.

Monteiro, M.R., Kugelmeier, C.L., Pinheiro R.S., Batalha M.O., and César A.S. (2018). Glycerol from Biodiesel Production: Technological Paths for Sustainability. *Renewable and Sustainable Energy Reviews*. Elsevier Ltd. doi:10.1016/j.rser.2018.02.019.

Nabi, M.N. (2010). Theoretical Investigation of Engine Thermal Efficiency, Adiabatic Flame Temperature, NOx Emission and Combustion-Related Parameters for Different Oxygenated Fuels. *Applied Thermal Engineering* 30, 839–844. doi:10.1016/J.APPLTHERMALENG.2009.12.015.

Pozarlik, A., Bijl, A., Alst, N. V, Bramer, E., and Brem, G. (2015). Pyrolysis Oil Utilization in 50 KWe Gas Turbine. In *18th IFRF Members' Conference – Flexible and Clean Fuel Conversion to Industry*, 1–10. Freising, Germany.

598 Rezzoug, S.A., and Capart, R. (2002). Liquefaction of Wood in Two Successive Steps: Solvolysis in
599 Ethylene-Glycol and Catalytic Hydrotreatment. *Applied Energy* 72, 631–44. doi:10.1016/S0306-
600 2619(02)00054-5.

601 Sallevelt, J. L.H.P., Gudde, J. E.P., Pozarlik, A. K., and Brem, G. (2014). The Impact of Spray
602 Quality on the Combustion of a Viscous Biofuel in a Micro Gas Turbine. *Applied Energy* 132,
603 575–585. doi:10.1016/j.apenergy.2014.07.030.

604 Seljak, T., and Katrašnik, T. (2019). Emission Reduction through Highly Oxygenated Viscous
605 Biofuels: Use of Glycerol in a Micro Gas Turbine. *Energy* 169, 1000–1011.
606 doi:10.1016/j.energy.2018.12.095.

607 Seljak, T., and Katrašnik, T. (2016). "Designing the Microturbine Engine for Waste-Derived Fuels."
608 *Waste Management* 47: 299–310. doi.org/10.1016/j.wasman.2015.06.004.

609 Seljak, T., Kunaver, M., and Katrašnik, T. (2014). Emission Evaluation of Different Types of
610 Liquefied Wood. *Strojniški Vestnik – Journal of Mechanical Engineering* 60, 221–31.
611 doi1.10.5545/sv-jme.2013.1242.

612 Seljak, T., Oprešnik, S.R., Kunaver, M., and Katrašnik, T. (2012). Wood, Liquefied in Polyhydroxy
613 Alcohols as a Fuel for Gas Turbines. *Applied Energy* 99: 40–49. doi:
614 10.1016/j.apenergy.2012.04.043.

615 Tzanetis, K. F., Posada, J.A., and Ramirez, A. (2017). Analysis of Biomass Hydrothermal
616 Liquefaction and Biocrude-Oil Upgrading for Renewable Jet Fuel Production: The Impact of
617 Reaction Conditions on Production Costs and GHG Emissions Performance. *Renewable Energy*
618 113,1388–1398. doi:10.1016/j.renene.2017.06.104.

619 Verma, Puneet, Mohammad Jafari, S.M. M.Ashrafur Rahman, Edmund Pickering, Svetlana
620 Stevanovic, Ashley Dowell, Richard Brown, and Zoran Ristovski. (2020). "The Impact of
621 Chemical Composition of Oxygenated Fuels on Morphology and Nanostructure of Soot
622 Particles." *Fuel* 259, 116-167. doi:10.1016/j.fuel.2019.116167.

623 Xu, D., Lin, G., Guo, S., Wang, S., Guo Y., and Jing, Z. (2018). Catalytic Hydrothermal Liquefaction
624 of Algae and Upgrading of Biocrude: A Critical Review. *Renewable and Sustainable Energy*
625 *Reviews* 97, 103–118. doi.org/10.1016/j.rser.2018.08.042.

626

Figure 1.JPEG

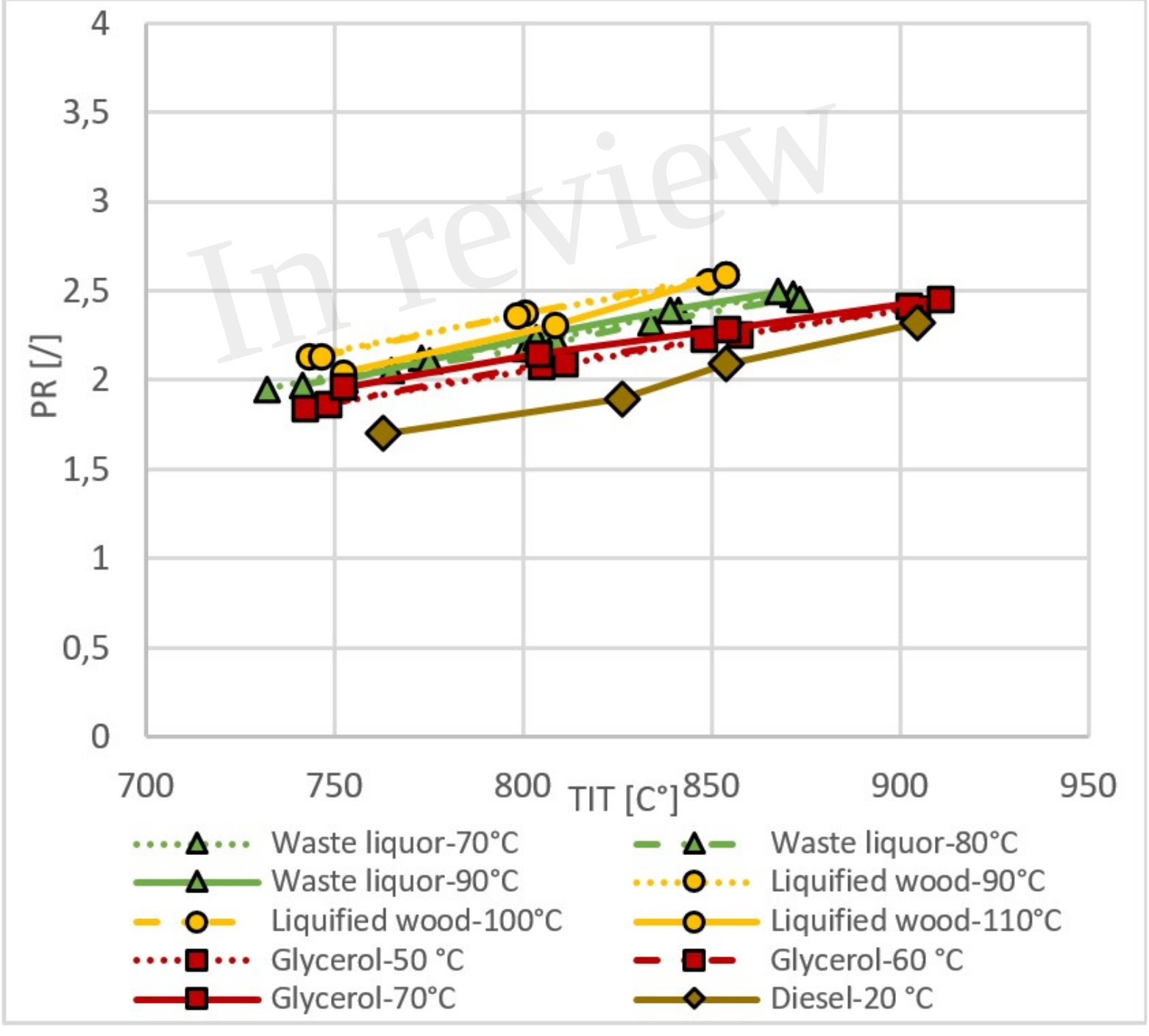

Figure 2.JPEG

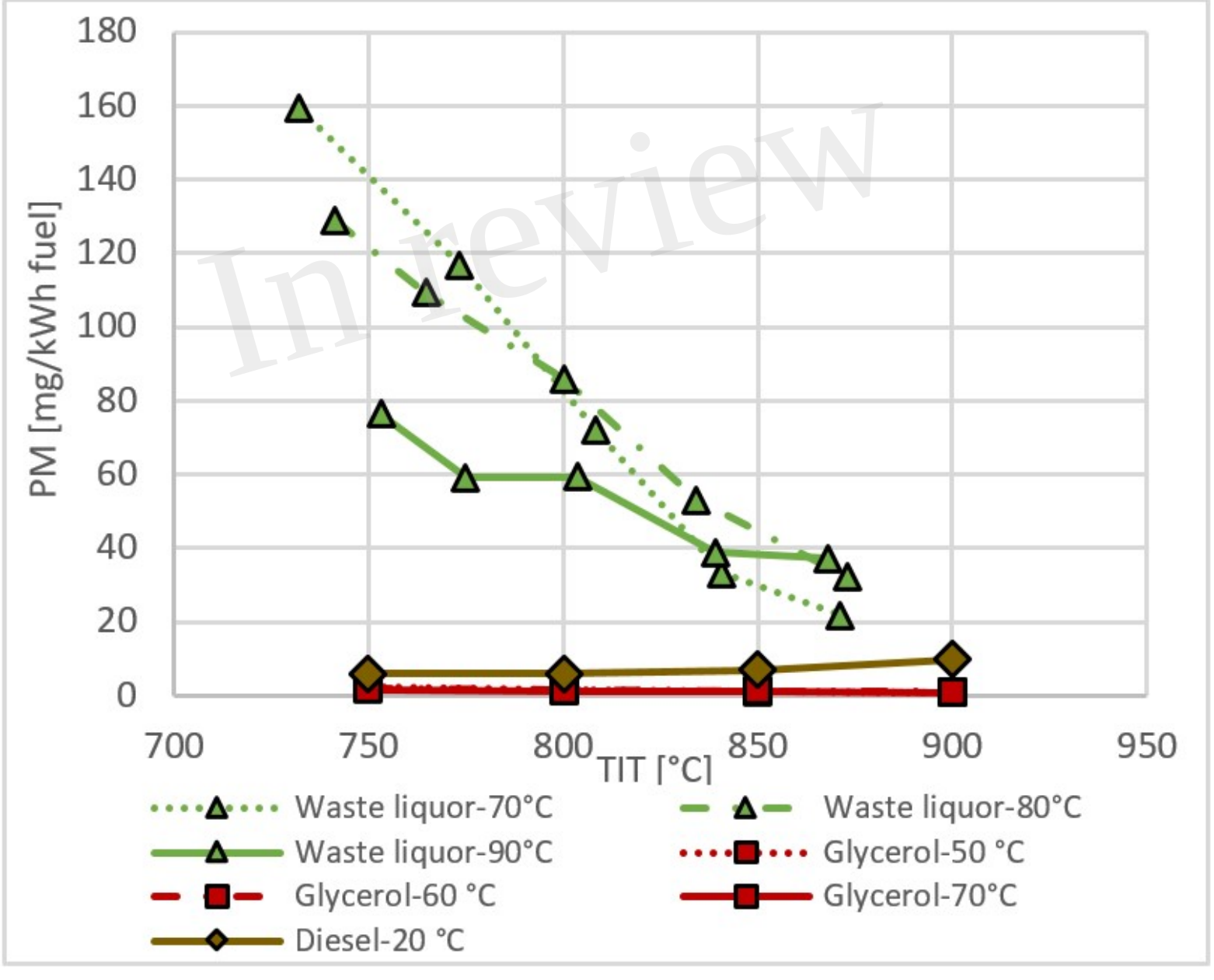

Figure 3.JPEG

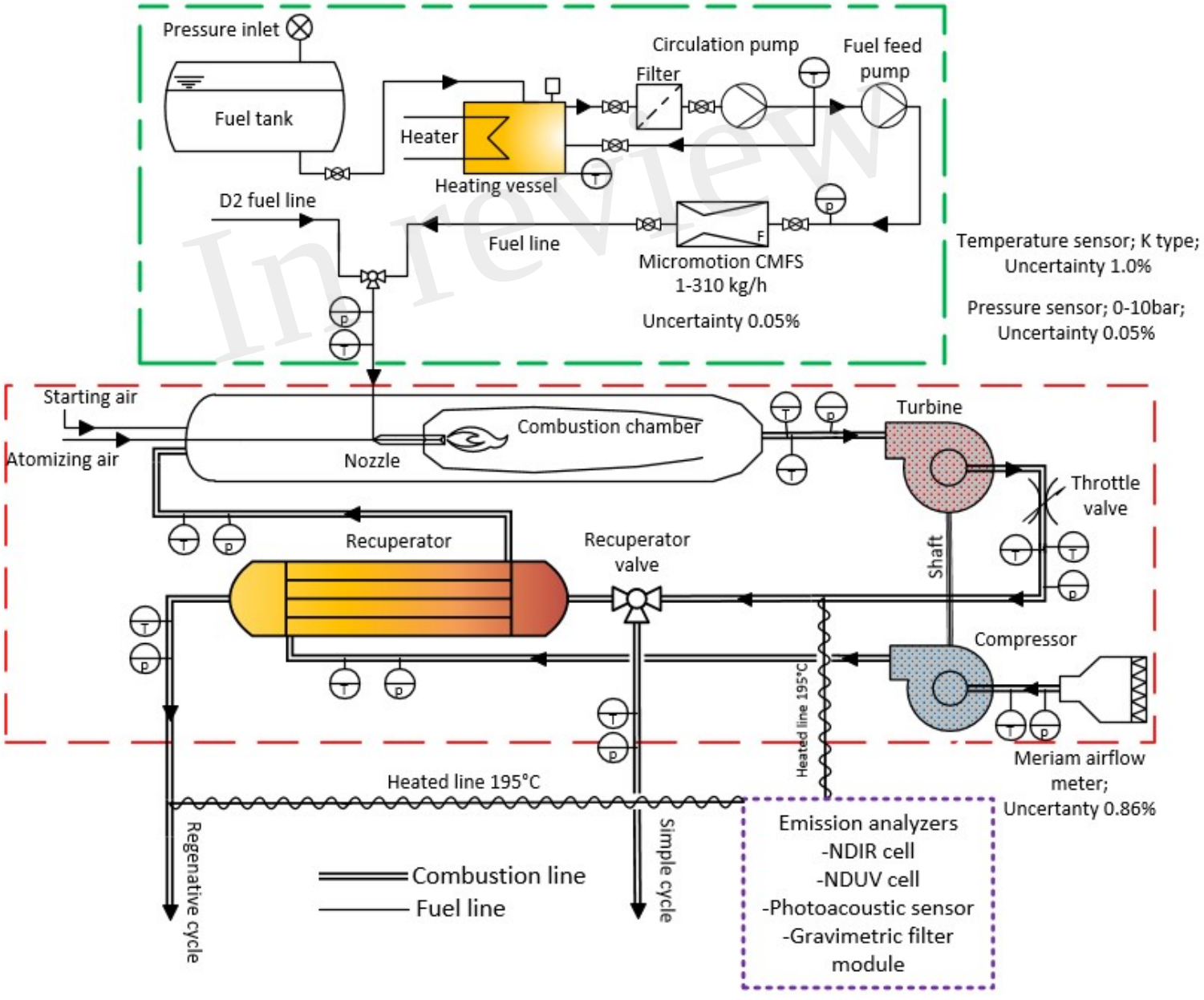

Figure 4.JPEG

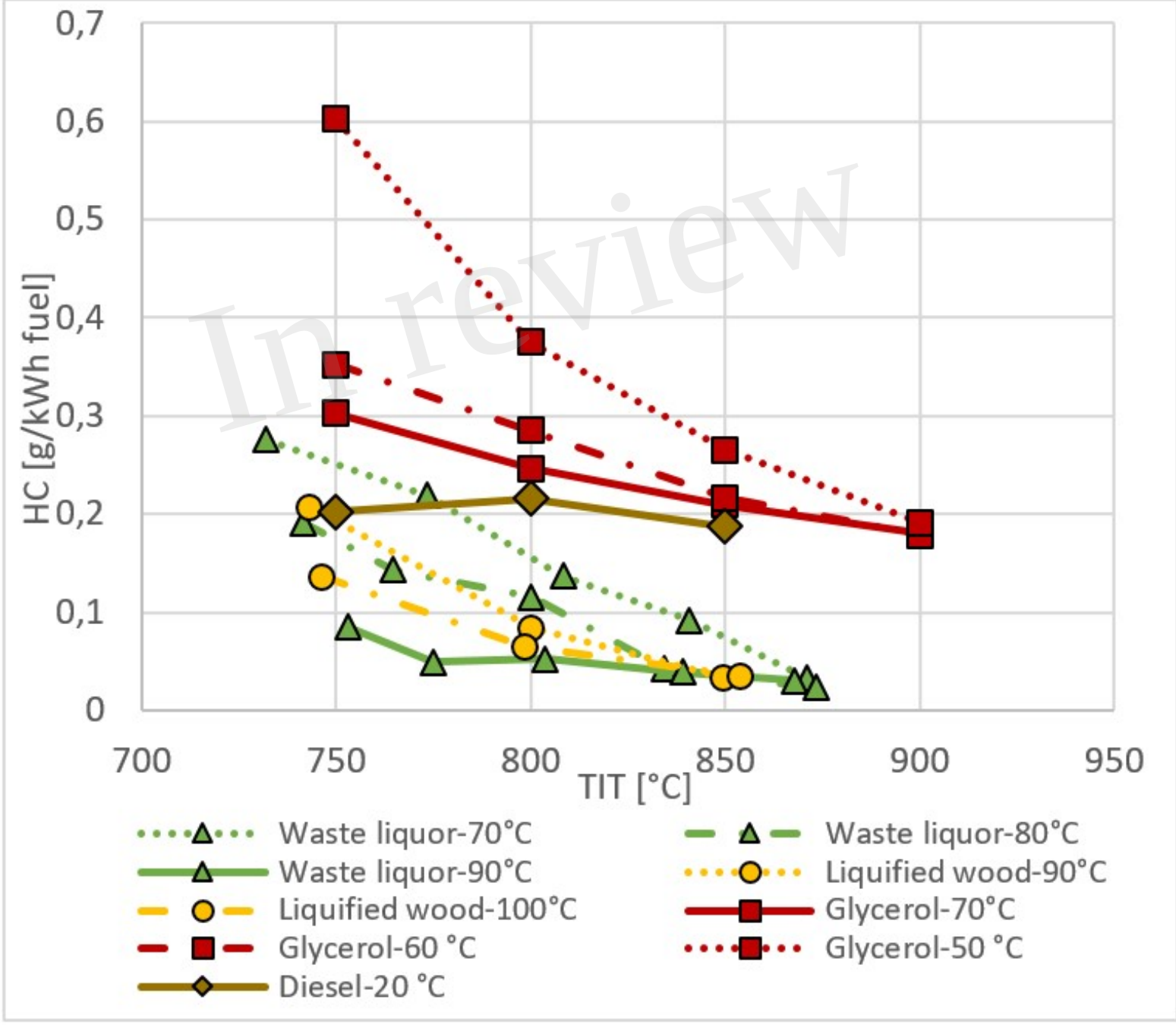

Figure 5.JPEG

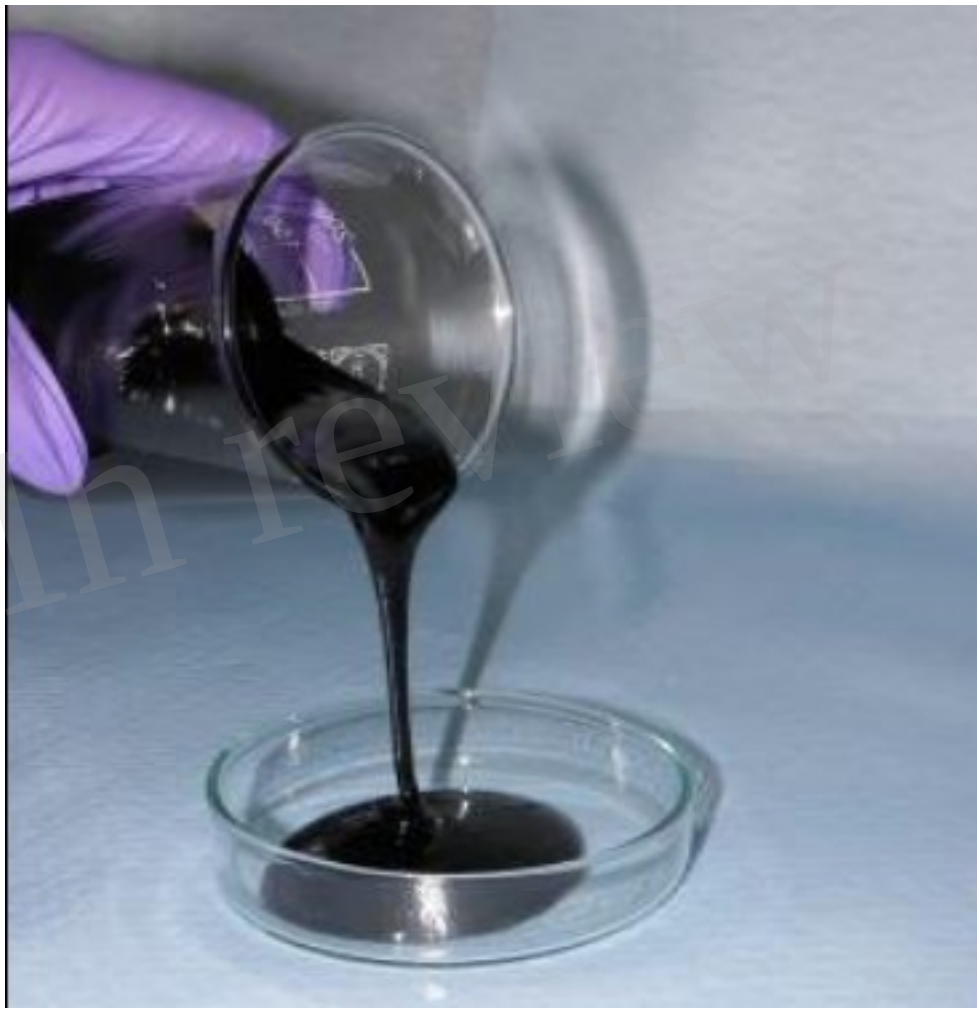

Figure 6.JPEG

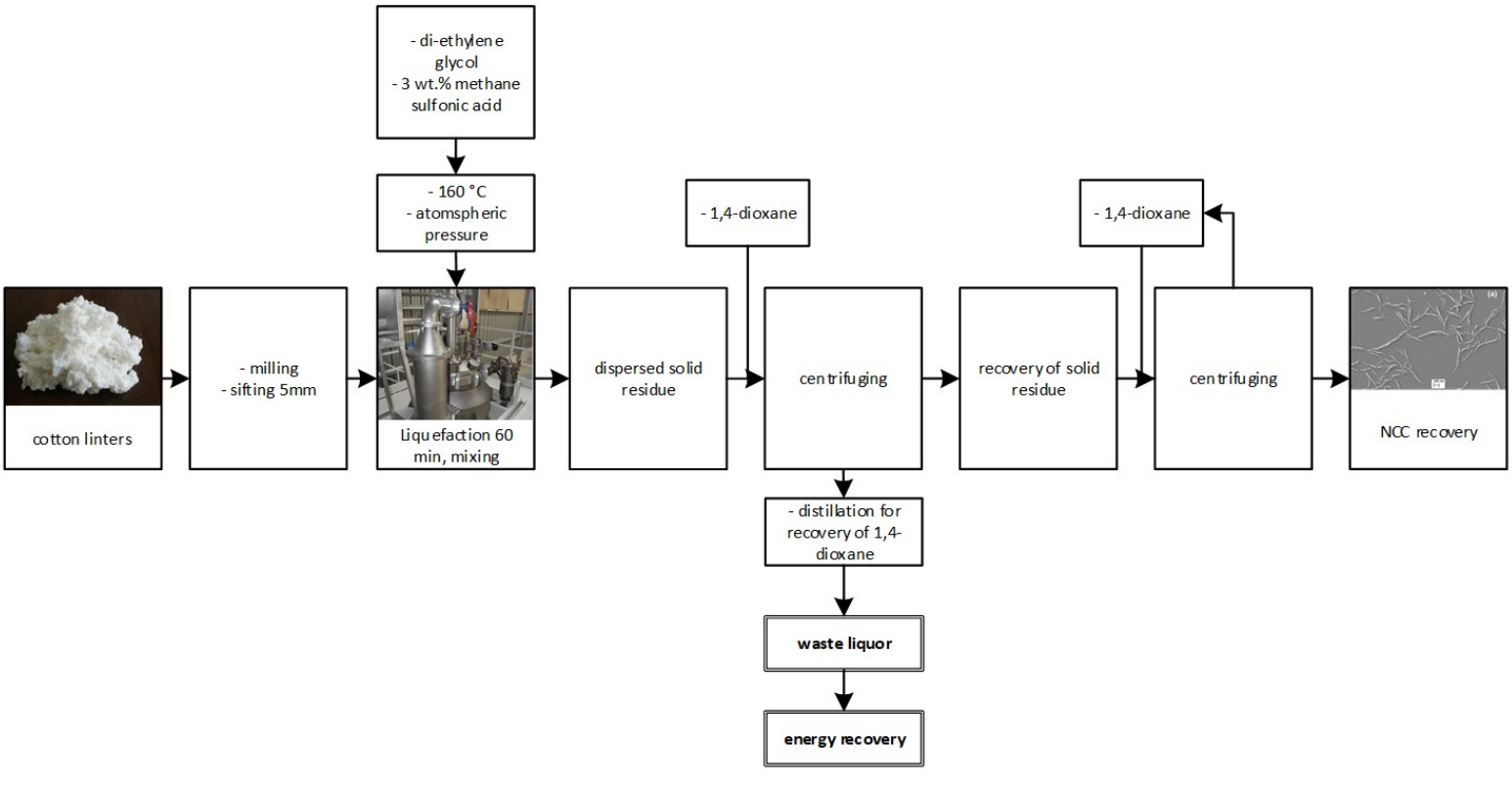

Figure 7.JPEG

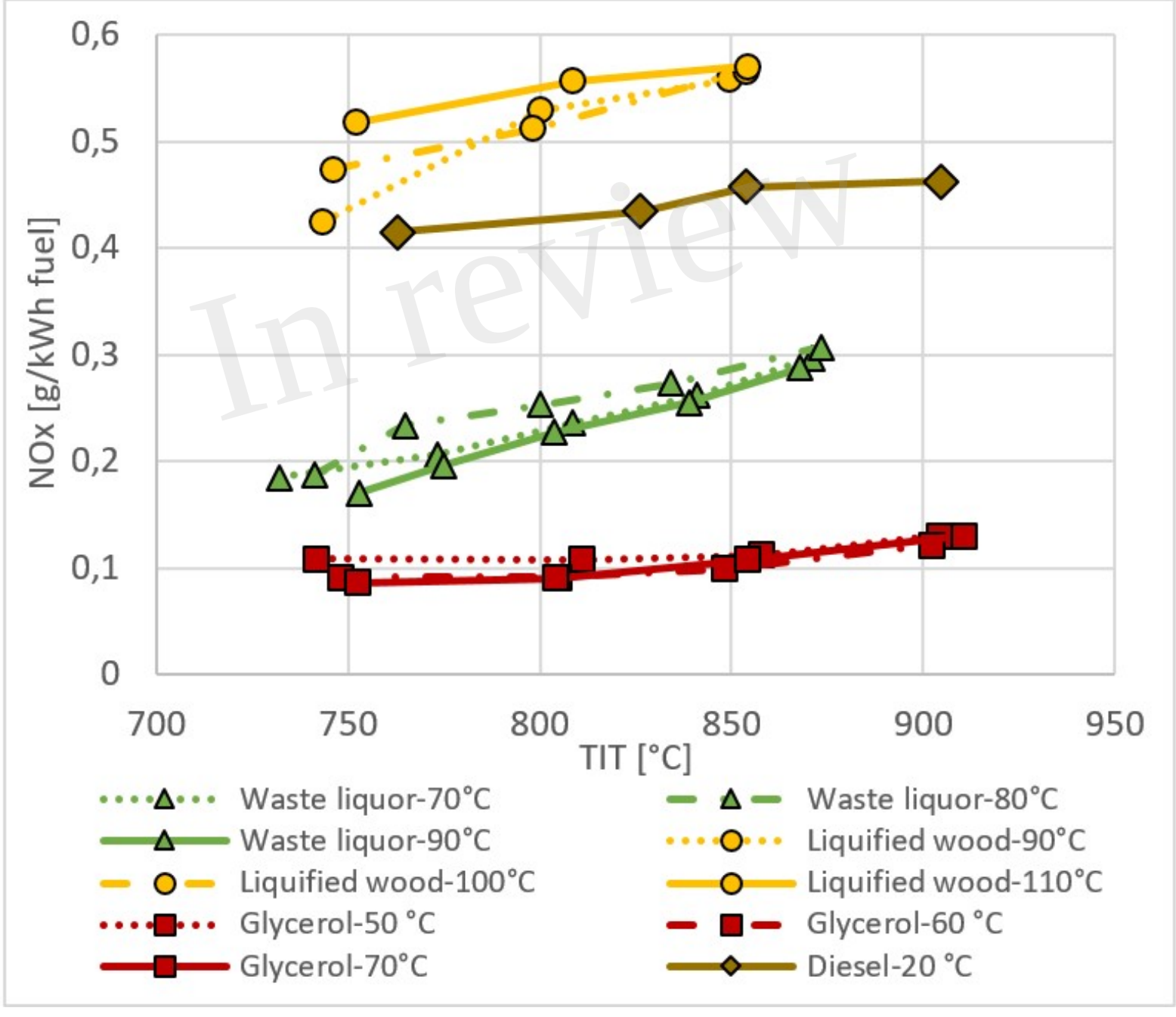

Figure 8.JPEG

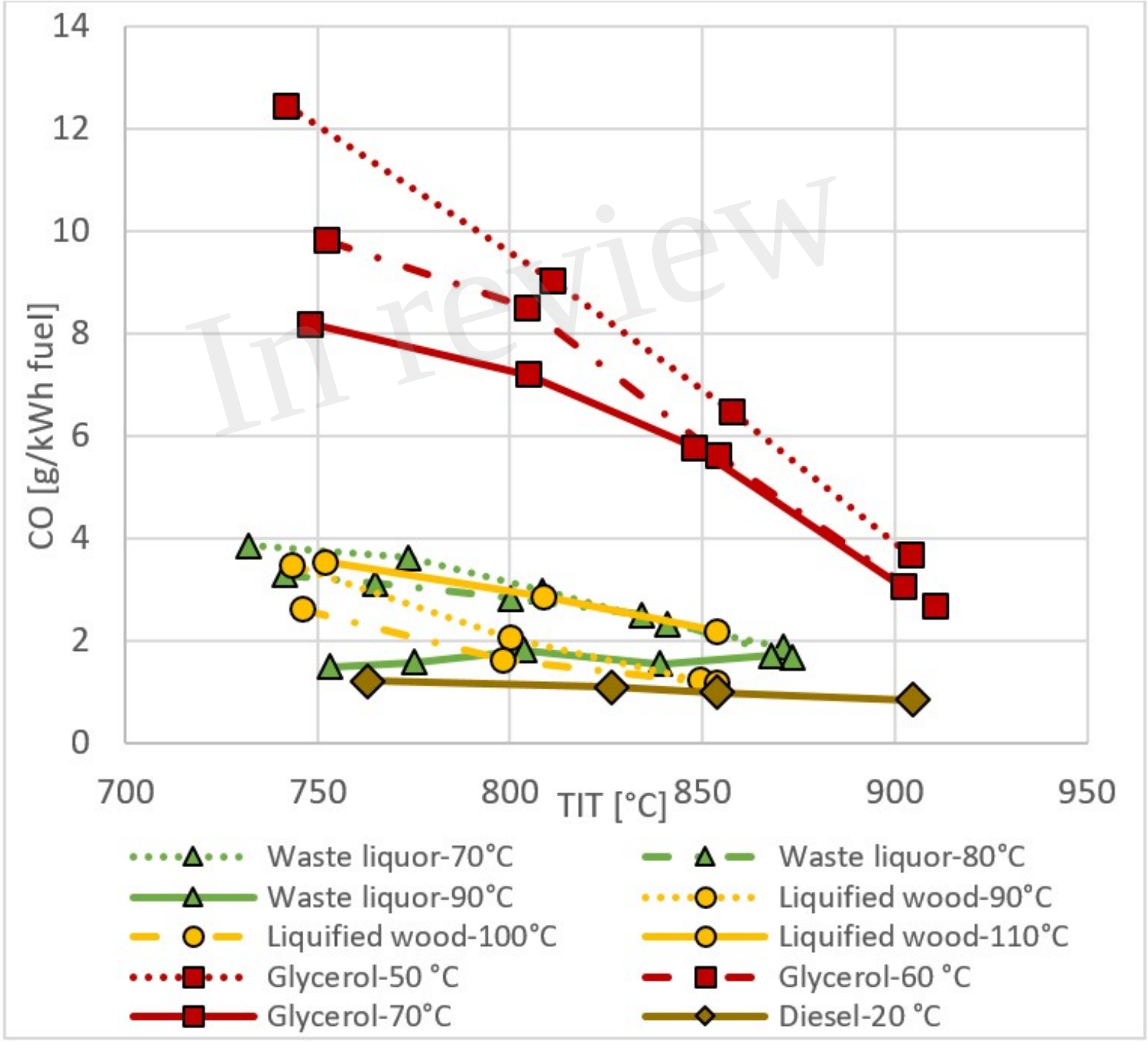

Figure 9.JPEG

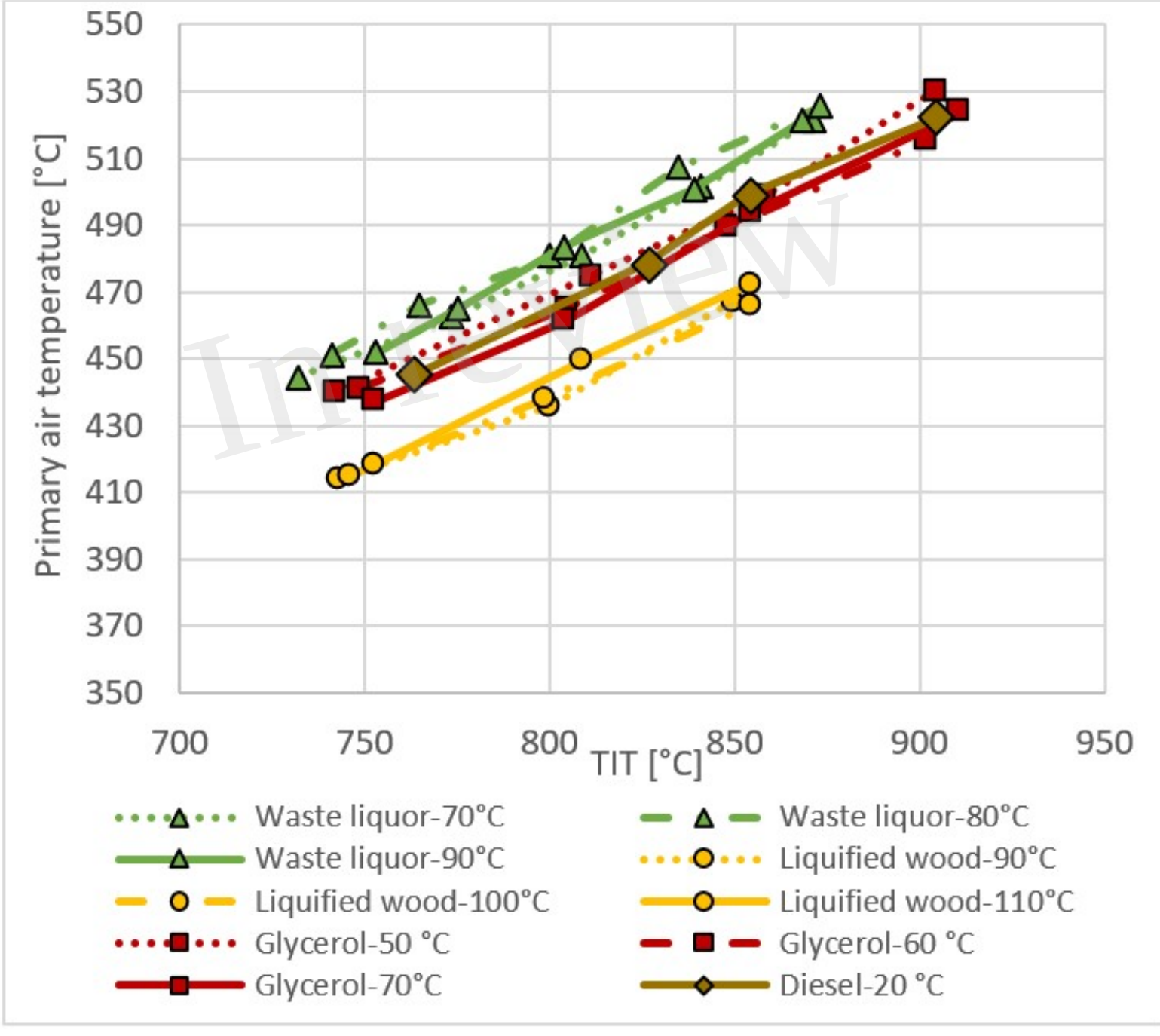

Figure 10.JPEG

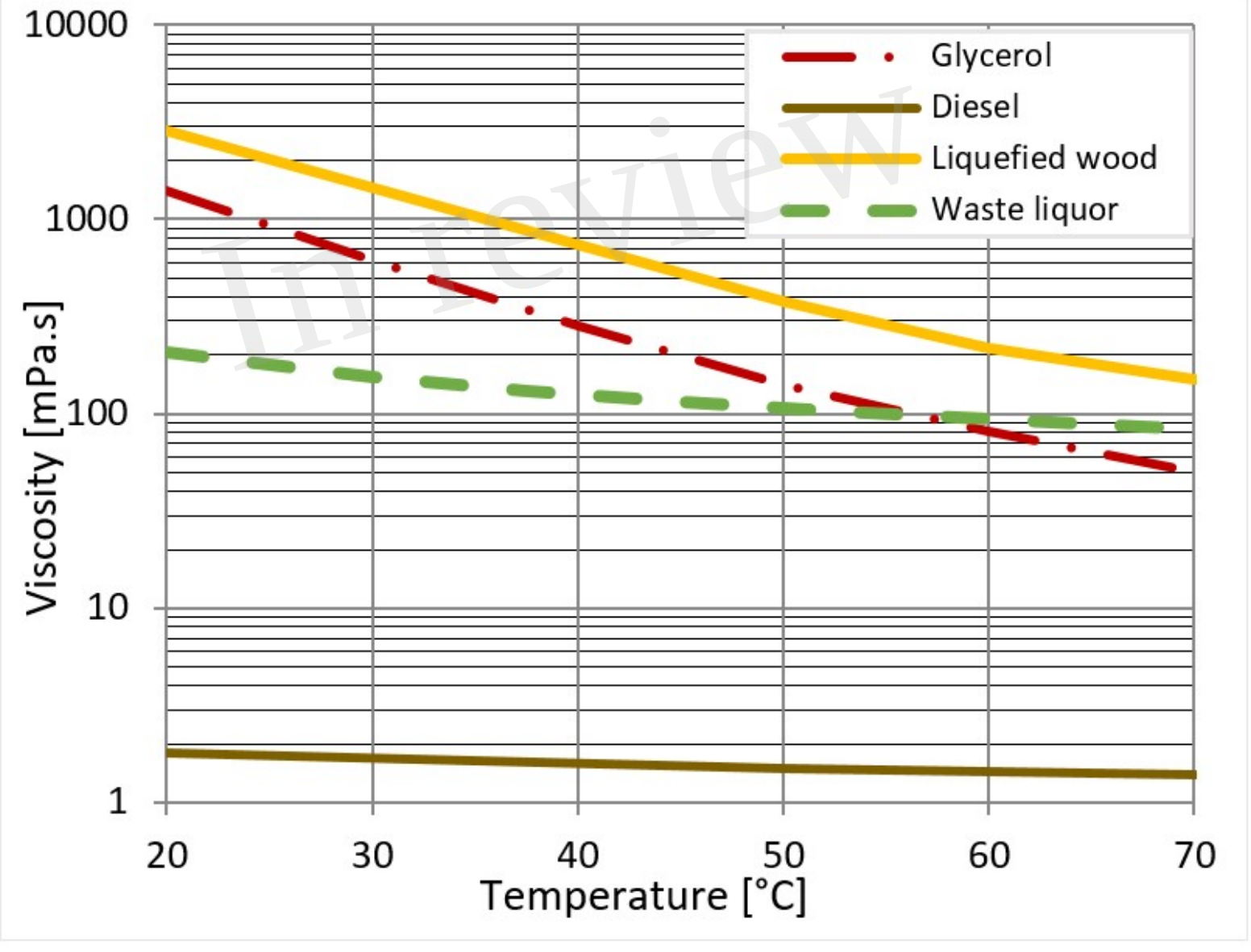

Figure 11.JPEG