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Understanding the effect of Intake temperature on the ϕ-sensitivity of toluene-ethanol reference fuels and neat ethanol

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Abstract

The low-temperature combustion (LTC) is an attractive concept that enables the modem combustion engines to move toward sustainability mainly by increasing the efficiency and decreasing the emissions. The modern combustion engines which are working based on the LTC concept have specific fuel requirements. Fuel ϕ-sensitivity is a key factor to be considered for tailoring fuels for these engines. Fuel with a high ϕ-sensitivity are more responsive to thermal or fuel stratifications; the auto-ignition properties of different air-fuel mixtures of these fuels, with different equivalence ratio (ϕ), are more diverse. This diversity provide a smoother heat release rate in stratified condition. In this study 11 different toluene–ethanol reference fuels (TERFs) in three research octane number (RON) groups of 63, 84, and 105 together with neat ethanol are evaluated. The Lund ϕ-sensitivity method is used to evaluate these fuels in a cooperative fuel research (CFR) engine. The effect of variation of intake temperature on pressure sensitivity of fuel at a constant combustion phasing is evaluated. This evaluation is performed at two intake temperature of 373 and 423 K, and the results are compared with the outcome of the Lund ϕ-sensitivity number with the intake temperature of 323 K. This study shows that the CR sensitivity response of different blends to the intake charge temperature variation depends on the fuel composition. Accumulated low temperature heat release and latent heat of vaporization. It proves that the fuel ϕ-sensitivity will vary under different thermodynamic conditions. There was a clear link between the accumulated heat released during the early reaction and CR sensitivity of the blends at different intake temperature of 373 and 423 K but the link with the latent heat of vaporization (HoV) found to be inexplicit.

Keywords: Homogenous charge compressed ignition; Intake charge temperature; Compression ratio; ϕ-sensitivity; Latent heat of vaporization.

Definitions/Abbreviations	
AcHR	Accumulated Heat Release
ATDC	After Top Dead Centre
CA50	Crank angle degree at which 50% of total Accumulated Heat is released
CFR	Cooperative Fuel Research
COV_{IMEP}	Coefficient of variation of Indicated Mean Effective Pressure
CR	Compression Ratio
DICI	Direct Injection Compression Ignition
DCR	Digital Counter Reading
GDI	Gasoline Direct Injection
HCCI	Homogeneous Charge Compression Ignition
HoV	Latent Heat of Vaporization
HRR	Heat Release Rate
HTHR	High Temperature Heat Release
ICE	Internal combustion engine
LTC	Low Temperature Combustion
LTGC	Low Temperature Gasoline Combustion
ITHR	Intermediate Temperature Heat Release
LTHR	Low Temperature Heat Release
MON	Motored Octane Number
OI	Octane Index
PFS	Partial Fuel Stratification
PPC	Partially Premixed Combustion
PRR_{Max}	Maximum Pressure Rise Rate
P_{SMC}	Pressure at Start of Main Combustion
RON	Research Octane Number
SI	Spark Ignition
S	Octane Sensitivity: RON-MON
SER	Start of Early Reactions
SMC	Start of Main Combustion
TAcHR	Total Accumulated Heat Release
TDC	Top Dead Centre
TERF	Toluene-Ethanol Reference Fuels
Tin	Intake Temperature
ϕ	Equivalence ratio

1 Introduction

The necessity of environmental protection and transition to sustainable society is not limited to a subject for scientific debates anymore. Every part of today's society from child and youth education to health care system is trying to be a part of this transition. Global warming is the most mentioned environmental challenge and carbon dioxide (CO_2) is the most known emission responsible for the global warming and climate change (1,2). Fig. 1 depicts the contribution of different sectors to the total CO_2 emissions. The data is extracted from IEA 2021 report(3). In 2019 the CO_2 emission share of transport sector has increased 2% compared to 1990, although 2% might not seem a significant increase, but since the total CO_2 emissions has 65 % increase from 1990 to 2019 the actual increase in CO_2 emissions of transport sector is 78% (3,4)! The accelerating increment in the numbers of privet vehicles, marine engines, airplanes, and heavy duty transportation are the reasons for this raise. It can be concluded that global transport sector is responsible for almost a quarter of total produced CO_2 during 2019 (5).

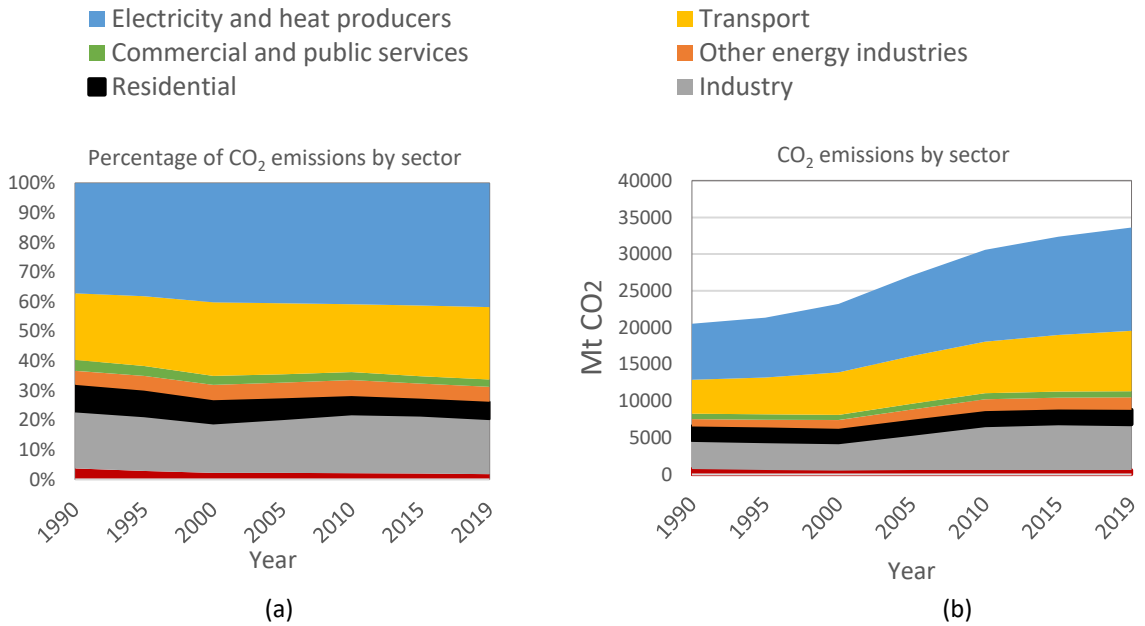

Fig.1 Global CO_2 emissions by sector from 1990 till 2019.(3)

Combustion engine is a developed and established technology which contributes largely to the transportation of goods and people. By looking at the mentioned statistics, it is expected to see

many combustion engine researchers searching for alternative combustion technologies to increase the engine efficiency and decrease the tailpipe emissions and move toward sustainable transport systems(6,7). There are some modern combustion technologies that have higher efficiency and lower emissions and working based on an alternative combustion concept known as low temperature combustion (LTC)(8–16). Like many other modern combustion technologies, the LTC engines, are working at lower temperature and higher pressure compared to the older engine generations (17–19). This working principle, makes the RON and MON inappropriate to completely predict combustion behaviour of in modern engines(17,20). Many researchers have developed different empirical or numerical methods to understand fuel behaviour in modern engines. The indexes such as the toluene number (21), octane index (17,22,23), HCCI fuel index (24), numerical ϕ-sensitivity evaluation method (25)and Lund-Chevron HCCI number (26) are more appropriate for modern engines and modern combustion concepts. Lund ϕ-sensitivity number (by the authors) is one of the empirical methods that evaluates and measures the ϕ-sensitivity property of different liquid fuels independent of fuels RON or MON (27).

The other side of the sustainable internal combustion engine (ICE) story is the fuel. Improving the technology of ICE alone will not solve the emission problems of ICE if these engines continue burning fossil fuels. The possibility of burning renewable fuels in both conventional and modern engine is another attractive area in ICE research. Many researchers have investigated the combustion of renewable fuels in both conventional and modern engines (7,11,28–35). Some researchers and companies moved one step further and have designed or manufactured special engines compatible with specific biofuels like bio-hydrogen (36,37). In fact, burning of fossil fuels is the main reason for the combustion engines to be considered unsustainable and when it comes to the renewable fuels the combustion behaviour of these fuels is not fully understood yet. This lack of knowledge is more pronounced when it comes to the combustion behaviour of renewable fuels in advanced combustion engine concepts like LTC. Homogeneous charge compression ignition (HCCI), is one of the most promising LTC technologies to increase fuel efficiency and decrease nitrogen oxides (NOx) and particular matters (PM) emissions. An HCCI engine merges two conventional combustion technologies of spark ignition (SI) and direct injection compression ignition (DICI) together. In an HCCI engine, the homogenous mixture of fuel and air enters the

combustion chamber or forms inside the combustion chamber, similar to a SI engine; the piston compresses the fuel-air mixture in the cylinder and the temperature increase during the compression leads to the auto-ignition of the mixture, like DICI combustion. HCCI combustion is widely used for fundamental fuel and combustion studies(24–27,38–40); because HCCI combustion is not affected from flame propagation like SI engines or diffusion flame like DICI engines. Sometimes, HCCI combustion is used by researchers as a platform to test fuels for further application of the results in other combustion technologies. As an example one of the most challenging combustion phenomenon is knock in spark ignition (SI) engines which limits the engine efficiency and even could damage the engine. By proper understanding of the heat release rate (HRR) and auto-ignition behaviour of homogeneous mixtures with different ϕ levels designing strategies for avoiding knock would be possible (41–43). Whenever the HCCI combustion is used as a platform to understand the fuel behaviour in the conventional ICEs, it is important to design the experiment based on the appropriate input parameters for the conventional ICEs (intake temperature and pressure, exhaust gas recirculation etc.). While, due to the different working principle of LTC combustion, the appropriate input parameters for the conventional ICE necessarily are not optimum for an HCCI engine. As an example the intake temperature of conventional engine is preferable to be the ambient temperature and in case of using exhaust gas recirculation (EGR) the cooled EGR is recommended to lower the nitrogen oxides (NOx) emissions (44–47). The most common and promising way for homogeneous charge preparation is preheating of air and fuel mixture before or in the intake manifold (19,48). Since the HCCI combustion is a type of LTC a high intake temperature (Tin) will not have a significant effect on the NOx formation and therefore would not be a limiting factor (42,49,50). The other methods for preparation of homogeneous mixture of an HCCI combustion are injection timing, using high EGR and negative valve overlap (NVO)(48,51–55). NVO method benefits from variable valve timing and traps the exhaust gases inside the combustion chamber. The NVO method increases the internal EGR and ensures that the exhaust valves are closed before the exhaust gases are completely discharged from the combustion chamber. In that way the initial in-cylinder temperature increases(56) .

The authors have explained in Lund ϕ-sensitivity methodology paper (27) that this method is an empirical method for ϕ-sensitivity evaluation of renewable fuels, conventional fuels, or any blends of conventional and renewable fuels. Alemahdi et.al. emphasis that the empirical Lund ϕ-sensitivity method can evaluate fuels which do not have a detailed kinetic mechanism(27). This method is based on the CR sensitivity of different fuels to the variation of ϕ in the intake temperature of 323 K .The following equation shows the way that the Lund ϕ-sensitivity number is calculated:

$$Lund\ \phi - sensitivity = -\frac{1}{CR}\frac{\Delta \mathrm{CR}}{\Delta \phi}$$

Charge preheating is a way of preparing a homogenous mixture for HCCI combustion. Applying hot EGR is another way to ensure mixing and increase the dilution level. Both these methods increase the charge temperature and as a result the fuel reactivity increases (17,25,40,43,57). This means that the pressure sensitivity of the fuel increases and that fuel auto-ignites at a lower pressure. How does this lower pressure and higher temperature of auto-ignition, influence the Lund ϕ-sensitivity number?

In this study the ϕ-sensitivity of different surrogate gasolines using the Lund ϕ-sensitivity method at a high intake temperatures of 373 and 423 K is evaluated to nail these objectives:

i. To study the impact of HoV on the ϕ-sensitivity of the blends.
ii. To evaluate the response of ϕ-sensitivity of the blends to the increased charge temperature because of charge pre-heating or hot EGR to prepare homogenous charge.
iii. To investigate the possible benefits of ϕ-sensitivity investigation at different intake temperature for fuel tailoring base on the desirable combustion concept.

2 Test methodology

In this study a cooperative Fuel research (CFR) engine with variable compression ratio (CR) is used to measure the effect of different intake temperatures on fuel's ϕ-sensitivity by measuring the Lund ϕ-sensitivity number of different fuels. A CFR engine is a standard engine with variable compression ratio which is primarily used to test research and motored octane numbers (RON

and MON). CFR engine can be used for fuels with different physical and chemical properties. The engine specification is presented in Table 1.

Table 1.CFR engine specifications.

CFR engine	
Parameters	**Value**
Displacement volume	612 cm^3
Number of cylinders	1
Bore	83 mm
Stroke	114 mm
CR	Variable (4:1 to 18:1)
Number of valves	2
Intake valve opens	10°ATDC ±2.5°
Intake valve closes	146°BTDC ±2.5°
Exhaust valve opens	140°ATDC ±2.5°
Exhaust valve closes	15°ATDC ±2.5°
Con. rod length [mm]	254
TDC clearance height	(114.3)/(CR - 1)
Head/bore area ratio	1
Piston/bore area ratio	1
Wrist pin crank offset	0

The modified CFR engine used in this study, presented in Fig. 2, is equipped with four port fuel injectors, which together with the intake air heater allows a homogeneous charge with an adjustable temperature and facilitate HCCI combustion of different fuels. The experiments is performed in a naturally aspirated condition. An intake air refrigerator unit is used to ensure constant intake air humidity.

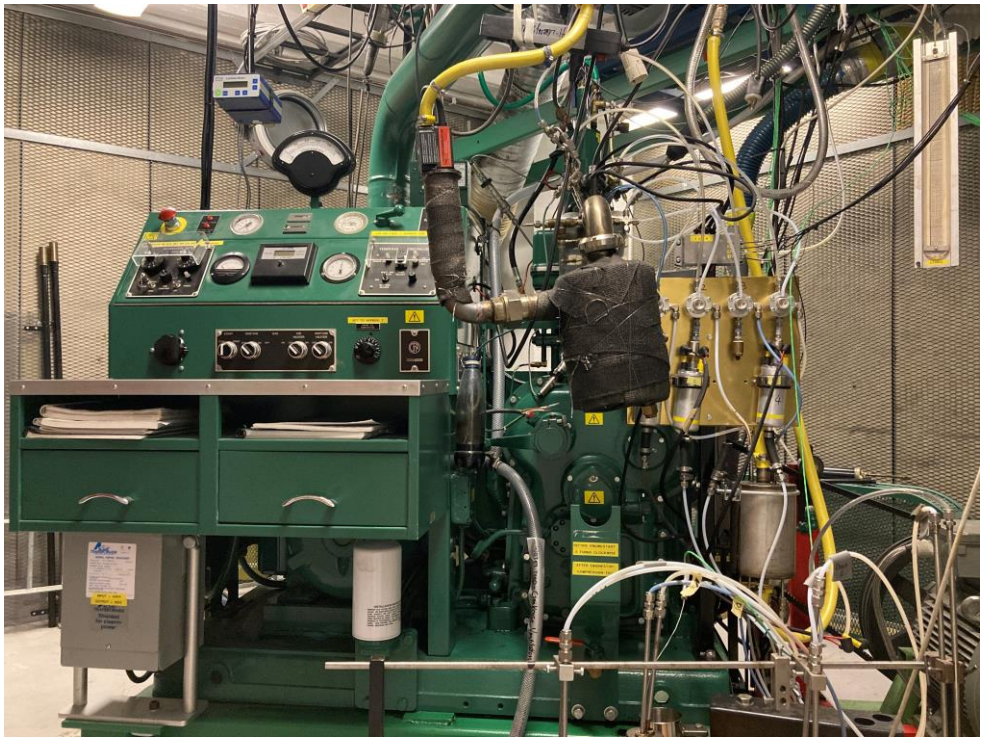

Fig.2. The modified CFR engine at the Energy sciences lab of Lund University.

The CR ratio of a CFR engine is adjustable. The cylinder head of a CFR engine moves upwards or downwards to increase or decrease the clearance volume and therefore varies the CR. An electrical motor is providing the required power for the movements of cylinder head.

Fig. 3 shows a schematic of the experimental apparatus. The inlet charge temperature is measured by a thermocouple mounted close to the inlet valve. The instantaneous in-cylinder pressure is measured using a Kistler piezoelectric sensor mounted inside the cylinder. At intake and exhaust manifold very close to the intake and exhaust valves, the time-averaged pressures were measured. The heat release calculation is based on Heywood (58). 300 successive combustion cycles as well as 100 successive motored cycles are sampled. For every test point the in-cylinder pressure measurements are filtered and averaged. The resolution for the instantaneous signals is 0.2 crank angle degree (CAD).

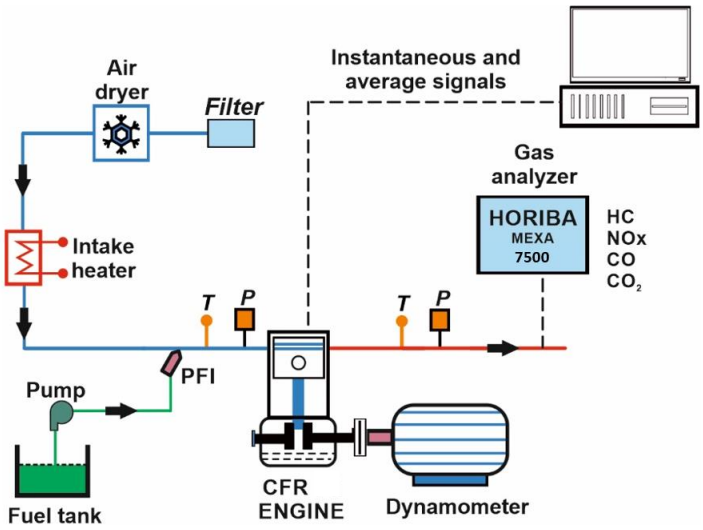

Fig. 3. Schematic of the modified CFR engine.

A Horiba Mexa 7500 analyzer system is used to measure NOx, unburned hydro carbons (UHC), carbon monoxide (CO), and carbon dioxide (CO_2). Oxygen and equivalence ratios are calculated from the exhaust gas emissions. In this study the HCCI combustion has been performed at four different levels of equivalence ratios, ϕ, constant rotation per minute (RPM) of 900 and intake temperatures (Tin) of 373 and 423 K. The details of experimental set up is presented in Table 2.

Table 2. Engine operating conditions.

Input parameters	Value	Variation
Intake Charge Temperature, Tin	373, 423 K	± 1 K
Pin	0.98 bar	± 0.03
RPM	900 RPM	± 2
Coolant Temp	373 K	± 1 K
Oil temperature	330 K	± 8.5 K
CA50	3 °CA	± 1 °CA
CR	Variable	-
Equivalence ratio ϕ	0.31, 0.33, 0.35, 0.37	± 0.005

The desirable combustion phasing of CA50 ≈ 3 is achieved by adjusting the compression ratio, CR and the ϕ is adjusted by variation of fuel mass.

2.1 Fuel selection

Due to the composition complexity of gasoline, surrogate fuel have been widely used in fundamental research (26,59–62). using surrogate fuels instead of gasoline helps researchers to

limit chemical and physical complexity of the tested fuels and make a base line for further experiment on renewable fuels (21,63). Single component, binary blends and multiple blends are various categories of surrogate fuels (64,65). PRF (Primary Reference Fuel) are blends of two paraffins, iso-octane and n-heptane which have been used to develop RON and MON test methods. PRFs are not octane sensitive, meaning that the RON and the MON for these blends are equal, RON – MON ≈ 0. Therefore, PRFs seems not to be a proper representative of gasoline and renewable fuels which all have octane sensitivity higher than 1. Therefore, in this study toluene-ethanol reference fuels (TERFs) have been selected and designed. Toluene is the most important aromatic components of gasoline and its content in gasoline can be up to 35% (65–67). The other component is ethanol which is widely used as a gasoline additive to enhance octane number and as a renewable fraction of fossil fuel (16,49,64,68). The blend components properties are presented in Table 3.

Table 3. Properties of blends components, iso–Octane and toluene data from (58), *n–heptane data from* (69), *and Ethanol Data provided by the project partner Preem.*

	Hydrocarbon class	Fuel	Molecular formula	RON	S= RON - MON	Boiling Point, K	Heat of Vaporization, KJ/Kg	Calorific Value MJ/ Kg	Oxygen content %
1	**Oxygenated**	Ethanol	C_2H_5OH	108	14	352	846	27	35
2	**Aromatics**	Toluene	$C_6H_5CH_3$	121	14	383	351	40.58	0
3	**iso-paraffins**	iso-octane	C_8H_{18}	100	0	372	305	44.46	0
4	**n- Paraffins**	n-heptane	C_7H_{16}	0	0	371.5	321	44.56	0

The TERFs of this study have been designed in three different RON groups of 108, 84, and 63 to be able to isolate the effect of ϕ-sensitivity from RON. The fuel design is done using an in-house regression model provided by (59) and the RON and MON is estimated. To measure the RON and MON precisely, the blends have been sent to the project partner, Saybolt AB Sweden for further evaluation. Detailed information is presented in Table 4.

Table 4. The Fuel matrix.

	Fuel	iso-octane, vol.%	n-heptane, vol.%	Ethanol, vol.%	Toluene, vol.%	Calculated RON*	Measured RON	RON-MON= S*	RON-MON= S[1]	HoV J/g
1	T20E40RFn10	30	10	40	20	108.3	105.5[4]	13.2	14.4[4]	536
2	Ethanol	0	0	100	0	-	108	-	14	846
3	T65RFn5	30	5	0	65	108.72	105.9[4]	14.31	9.3[4]	340.5
4	E50RFn12	38	12	50	0	108.8	105.3[4]	12.1	14[4]	591
5	T25E30RFn40	15	40	30	25	84.52	85.4[4]	6.9	7.8[4]	490
6	E38RFn43	19	43	38	0	84.4	84.4[3]	5.4	5.7[3]	532
7	T50RFn30	20	30	0	50	82.7	83.8[2]	8.5	7.6[2]	334
8	PRF84	84	16	0	0	-	84	-	0	307
9	T10E10RFn49	31	49	10	10	63.0	63.1[4]	4.0	5.1[4]	376
10	E20RFn55	25	55	20	0	63.7	63.2[4]	4.2	4.9	432
11	T40RFn48	12	48	0	40	63.3	63.7[2]	6.9	5.7	332.9
12	PRF63	63	37	0	0	-	63	-	0	310

**Calculated based on a model introduced in* (59)
1 based on the measured RON and MON from different references.
2 (63)
3 (59)
4(measured by project partner, Saybolt AB Sweden)

As an example of blending, T10E10RFn49 is a blend, consisting 10% toluene, 10% ethanol and 49% n-heptane the iso-octane is used as the filler therefore in this case 31% iso-octane has been used. All the blending ratios are volumetric ratios. In some cases, the measured RON and MON are slightly different compared to the estimated values. The measured values were used in the data analysis of this paper.

For all fuels in this study the experimental input parameters are similar, as it is presented in Table 2. Then the CR is adjusted for each fuel according to the ϕ to maintain the CA50 constant and Cr is used as the main output to measure the Lund ϕ-sensitivity number of each fuel according to Alemahdi et al., (27) .

2.2 Different phases of HCCI combustion

Undesirable auto-ignition might damage the engine, like knock or super-knock in an SI engine. A controlled auto-ignition is desirable since the auto-ignition is the onset of HCCI or DICI combustion. HCCI combustion is a consequence of spontaneous auto-ignition of fuel-air mixture in a pressurized combustion chamber through an array of kinetics reactions. The initiation of significant reactions is extremely temperature dependent and occurs at temperatures higher than 550K (19). In two stage combustion the main chain branching exothermic reaction pathway of alkyl radicals (R) and (H_2O) with O_2 are the early reactions which are responsible for low temperature heat release (LTHR) so called low temperature reactions (LTR) (39,70,71). Intermediate temperature heat release (ITHR) is caused by the early reactions before the start of the main combustion and after the LTHR reactions also known as the intermediate temperature reactions that in two stage combustion occur after NTC regime. The main exothermic combustion reactions responsible for high temperature heat release (HTHR) are $H_2O_2 + M = OH + OH + M$, where M is called the third body and could be any molecule in the mixture. Fig. 4 illustrates the criteria and definition of distinct phases of combustion for HCCI combustion in this study adapted from (27). In this research based on (27), the point at which the rate of heat release exceeds 0.2 J/CAD and does not drop lower than this value is selected as the start of auto-ignition or start of early reaction (SER) point. The first point after NTC region that the rat of heat release has reached 5J/CAD is selected as the start of high temperature heat release (HTHR) and therefore main combustion.

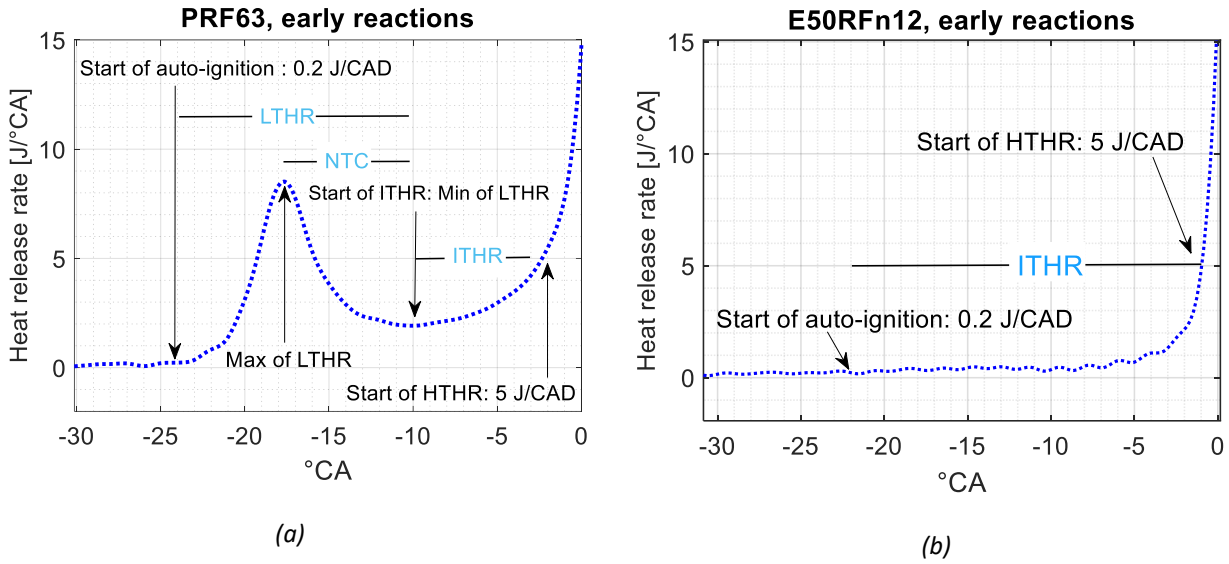

Fig.4. CAD selection for start of LTR and HTR: a) HRR during the early reactions for two-stage combustion; b) HRR during the early reactions for single-stage combustion

3 Results and discussion

In this section the heat release profile of different blends is investigated. Lund ϕ-sensitivity number at the intake temperature of 423K is calculated and the results are compared with the Lund ϕ-sensitivity number of the blends at intake temperature of 323K. Error bars in the plots of this study are the representative of the standard deviation of each parameter from the mean value.

3.1 HCCI Combustion and heat release behaviour of different blends:

HCCI combustion is a consequence of spontaneous combustions of fuel-air mixture in a pressurized combustion chamber. The Study of the rate of heat release (HRR) versus crank angle degree (CAD) gives us an inclusive overview of combustion phenomenon inside the cylinder. Mostly, it is straight forward to detect the existence of low temperature heat release (LTHR) by looking at the HRR plots. Other information like combustion duration, peak of HRR, CAD of start of combustion are also harvested from the HRR evaluation.

It has been frequently discussed in literatures that higher inlet temperatures (Tin) increases the octane reactivity of a blend (17,25,40,43,57,67,72) - meaning that the fuel will auto-ignite at a

lower pressure. The aim of this section is to explain how the higher Tin affects the Lund ϕ-sensitivity number, which is a measure of the fuel ϕ-sensitivity property of a fuel (27). From here in this paper and onward whenever ϕ-sensitivity is mentioned or discussed refers to the fuel ϕ-sensitivity property measured using Lund ϕ-sensitivity number (27).

In this section, HRR of one blend from each group, as the representative of that group is presented and discussed. The three selected blends contain all the four components that are discussed earlier, oxygenated, aromatics, iso-paraffins and n-paraffins. Fig. 5 depicts the HRR versus CAD for these three blends and pinpoints the start of early reactions and the main combustion for each blend at three Tin of 323, 373 and 423 K.

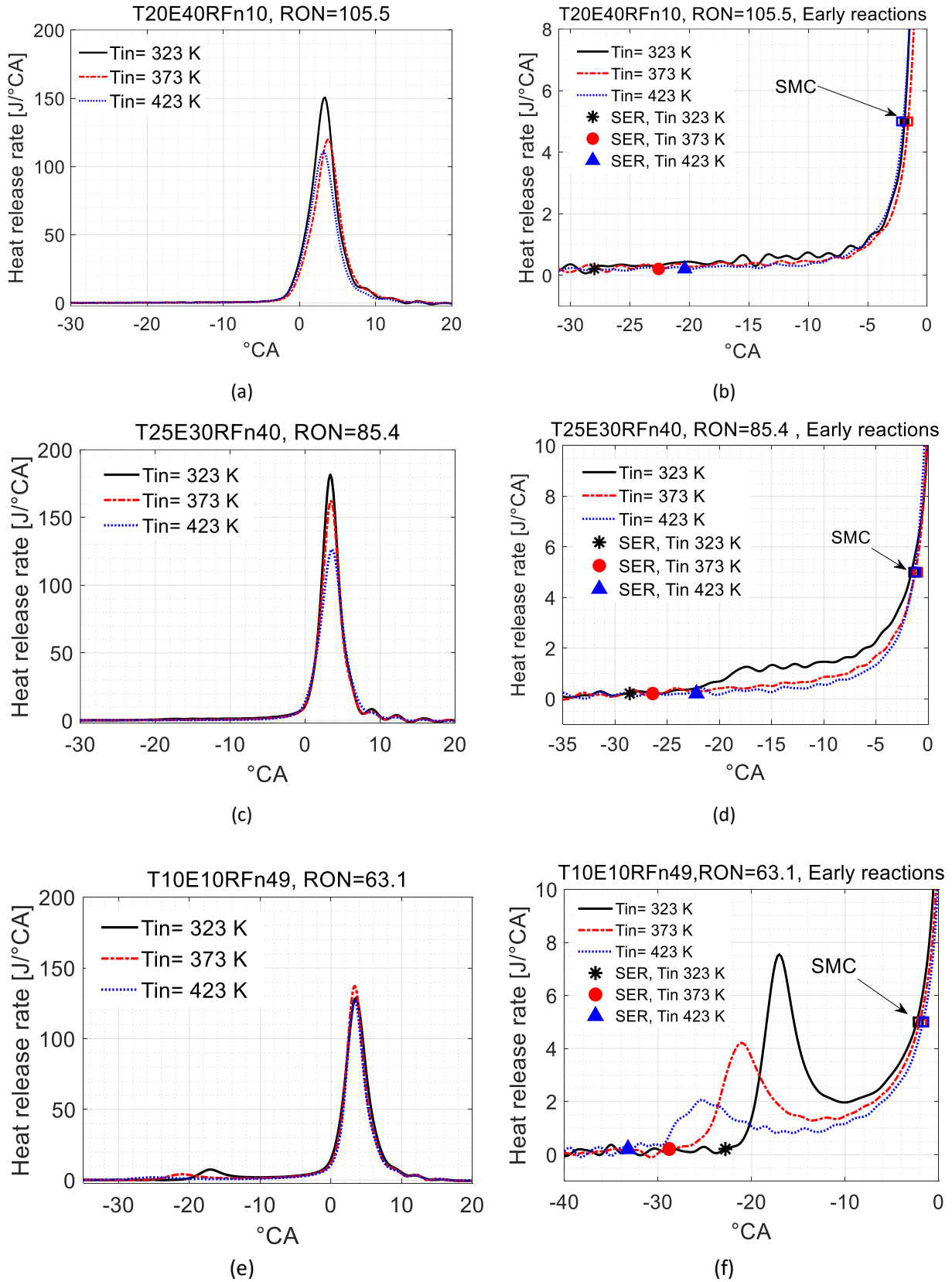

Fig 5. HRR versus CAD for TERF of each RON group at three intake charge temperatures.

Fig. 5 a-d show, for the blends with single stage combustion, the Ignition delay decreases with increase in Tin. At higher intake temperature (Tin) lower CR is required to maintain the CA50 constant. Lower CR decreases the in-cylinder pressure and leads to later start of combustion. On the other hand, for the blends with two stage combustion, increasing the Tin, increases the ignition delay (Fig. 5 e and f) by advancing the low temperature combustion. Low temperature reactions are more temperature sensitive than the main combustion reactions and the onset of low temperature reactions occurs at a lower temperature and pressure than the main combustion reactions (42,49,73,74). The main combustion reactions are happening at a temperature around 1000 K (19) while the low temperature reactions are initiating at around 550 K (48). An increase of intake temperature decreases the air mass entering the combustion chamber and therefore less fuel mass is needed to form a specific mixture strength compared to a lower intake temperature that the air density is higher. This thermodynamic condition affects the total energy that is released during the combustion; as a result, lowers the peak of HRR. The thermodynamic condition impact on the required fuel mass, is not the only reason for lower HRR at a higher Tin; the other reason is that the CR is adjusted to achieve a constant combustion phasing of CA50 ≈ 3. The higher Tin increases the blends reactivity (25,75–77) and therefore lowers the required CR to keep the CA50 constant. The lower CR together with the lower energy content of the fuel-air mixture, decreases the HRR as well as the total accumulated heat release (TAcHR). This reduction in TAcHR also affects the ratio of heat release during the early reactions to the TAcHR. This ratio is the main confirmed reason for the fuel ϕ-sensitivity property and therefore Lund ϕ-sensitivity number (25,27,78)

Figure. 6 displays the ratio of AcHR during the early reactions to the TAcHR for Tin 423K and ϕ≈0.31. The accumulated heat release during the early reactions is known as an indicator of fuel's ϕ-sensitivity (25,27,28,42). These studies emphasis on the direct correlation between the percentage of AcHR during the early reactions (both low and intermediate reactions) and the ϕ-sensitivity of a fuel. Those fuels that generate more accumulated heat during the early reactions have higher ϕ-sensitivity. Fig. 6 shows that the blends with ethanol content have significantly lower heat release during the early reactions. The quenching effect of Ethanol on LTHR is well known through experimental and numerical studies (49,73,79–82). This scavenging effect is due

to the consumption of $O\dot{H}$ and $H\dot{O}_2$ radicals, forming RCHO and HO_2 which are greatly stable at low temperature (31,41,82–84).

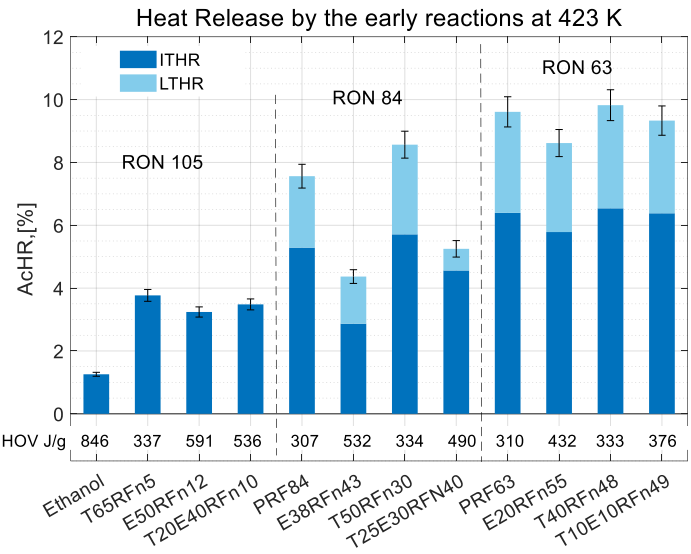

Fig.6. The ratio of AcHR during the early reactions over the TAcHR at Tin 423K for ϕ= 0.31.

The quenching effect of Ethanol on LTHR is well known through experimental and numerical studies (49,73,79–82). This scavenging effect is due to the consumption of $O\dot{H}$ and $H\dot{O}_2$ radicals, forming RCHO and HO_2 which are greatly stable at low temperature (31,41,82–84).

There is also a clear link between latent heat of vaporization (HoV) of the blend and the percentage of released heat during the early reactions. As it was expected the higher HoV, which in this case is a result of higher ethanol content in the blends, lowers the LTHR.

3.2 ϕ-sensitivity calculation according to the Lund ϕ-sensitivity number

In this section the method and the formula for the Lund ϕ-sensitivity number calculation, by the authors (27), is used to calculate the ϕ-sensitivity of the blends at Tin 423 K, considering this fact that the intake temperature for Lund ϕ-sensitivity number is meant to be 323 K. The objective of this calculation was to understand the ϕ-sensitivity concept under different intake temperature regimes and study the effect of higher intake temperature on Lund ϕ-sensitivity number and

therefore ϕ-sensitivity property of different fuels. The intake temperature of 423 K is selected because it is above the boiling point of all of blends components. The following equations (Eq.1 and 2) demonstrate the way that Lund ϕ-sensitivity number (Normalized CR) and corresponding normalized in-cylinder pressures at the start of main combustion are calculated (27):

$$\text{Lund } \phi - \text{sensitivity number} = -\frac{1}{CR}\frac{\Delta \text{CR}}{\Delta \phi} \qquad \text{Eq.1}$$

$$\text{Norm } P_{SMC} = -\frac{1}{P_{SMC}}\frac{\Delta P_{SMC}}{\Delta \phi} \qquad \text{Eq.2}$$

Since a higher Tin increases the fuel reactivity a lower CR (pressure) is required to maintain the CA50 constant. Fig. 3. shows the required CR to maintain the CA50 constant for two blends of the low RON group. Fig. 5 clearly depicts that increasing the intake temperature (moving from solid lines to dashed and doted lines) decreases the required CR to maintain the CA50 constant.

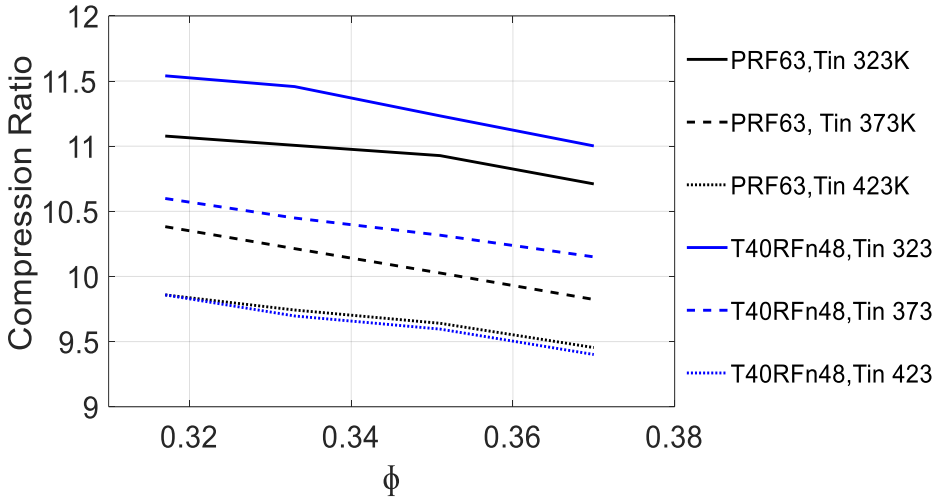

Fig. 3. Required CR for two blends (same RON) at different Tin.

The bar chart in Fig. 8 exhibits the normalized CR (Lund ϕ-sensitivity number) and the normalized in-cylinder pressure at the start of main combustion. This bar chart shows that for the RON≈105 and RON≈84 groups the pressure variation at the start of main combustion (P_{SMC}) follows the same trend as the variation in required CR to keep the CA50 constant dose.

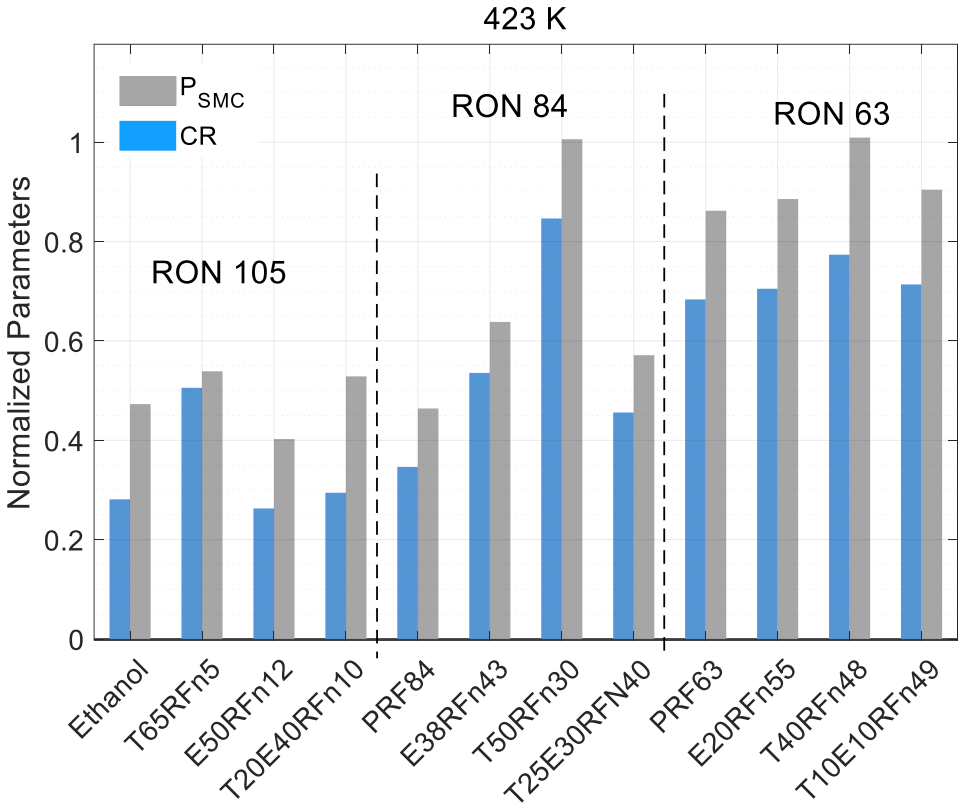

Fig. 8. Normalized CR and pressure at the start of the main combustion.

By contrast, for the RON ≈ 63 group this is not the case and the pressure variation at the start of main combustion (P_{SMC}) has the opposite trend compared to the variation in normalized CR.

Fig. 9 shows the Lund ϕ-sensitivity numbers at 323 and 423 K for the evaluated blends in this study. Ethanol and T65RFn5 are not included in this figure, because those fuels did not burn at the Tin ≈ 323K at the CR range of the CFR engine. As Fig.9 suggests, 6 out of 10 blends has significant change in the Lund ϕ-sensitivity number due to the intake temperature variation; for the other four blends (T20E40RFn10, PRF84, E20RFn55, and T40RFn48) the variations are within the standard deviation range and cannot statistically consider as significant variations. Among those 6 blends two of them show a decrease in Lund ϕ-sensitivity number at Tin≈423 K compared to Tin≈323 (T25E30RFn40 and T10E10RFn49). This behaviour suggests that these two TERFs are less ϕ-sensitive at high intake temperature. The main conclusion from Fig. 9 is that the TRFs have

the highest Lund ϕ-sensitivity number at Tin 423 while for the Tin 323, TERFs have the highest Lund ϕ-sensitivity number.

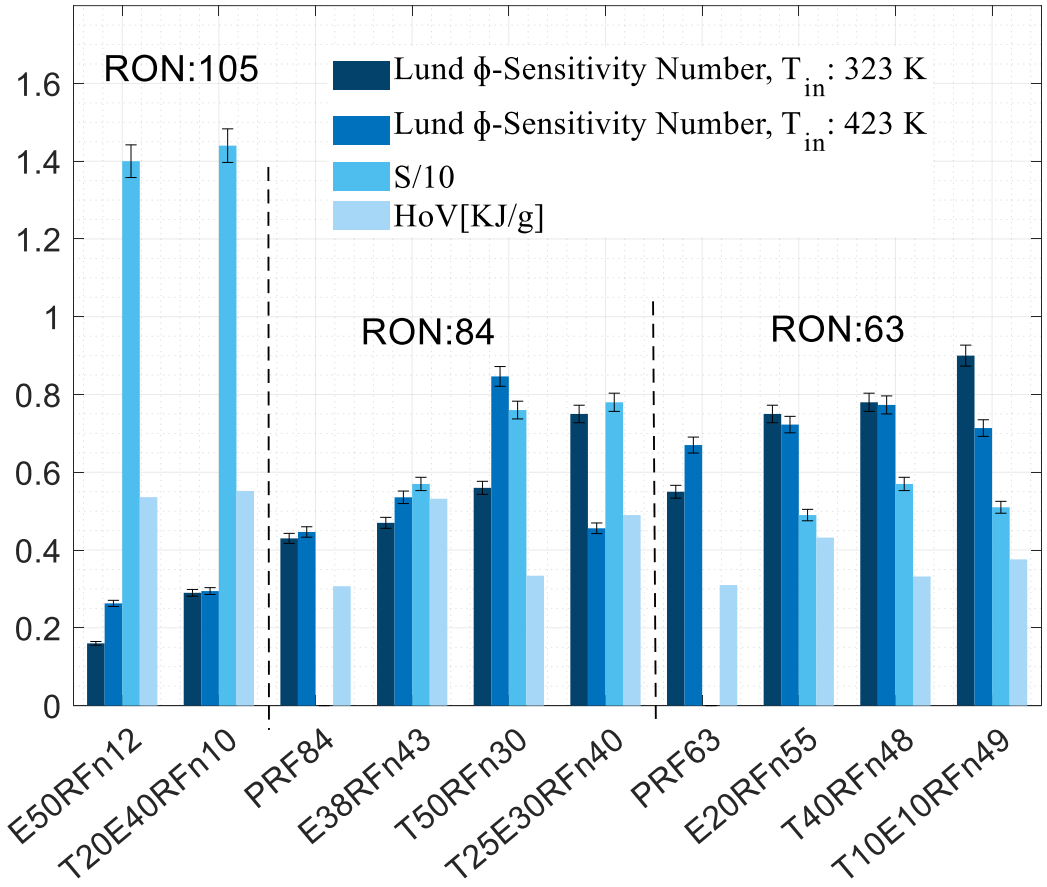

Fig.9. Lund ϕ-sensitivity number of the blends at two Tin 323 and 423K.

Fig.6 shows that TRFs at 423 K have the highest percentage of heat release during the early reactions compared to the other blends of their RON group. Which explained the reason for the higher fuel ϕ-sensitivity (25,27,85,86). The other point is that TEFRs show decrease in Lund ϕ-sensitivity number at Tin 423 compared to Tin 323. It means that the TERFs despite the other blends, in case of stratification inside the combustion chamber exhibit more inconsequent combustion at Tin 423. The chemical kinetics of the auto-ignition of toluene is discussed in different studies (21,41,42,59,72,80). Adding toluene to a fuel delays both the low temperature reactions and the main combustion (82,87). The reason for this effect is that toluene inhibits

certain intermediary species and acts as a radical sink. Toluene consumes very reactive radicals of $O\dot{H}$ and $\dot{H}$ by the unreactive benzyl radicals. In a parallel set of reactions, $H\dot{O}_2$ radicals produce H_2O_2. The H_2O_2 stays until the thermodynamic condition for the H_2O_2 decomposition is prepared. At this step, the H_2O_2 decomposition reactions terminates the toluene radical consumption reactions and initiates the main combustion reactions. That leads to a reduction of $O\dot{H}$ radical concentration and a longer presence of H_2O_2, which delays both cool flame formation and the start of the main combustion. This motive is the most important reason for the TERFs to have higher Lund ϕ-sensitivity number at an intake temperature of 423K compared to Tin 373 K. For the same reason TRFs have the highest Lund ϕ-sensitivity number in each RON group of this study, figure.5.

For T40RFn48 blend (RON≈63) Lund ϕ-sensitivity number variation due to the Tin variation is not significant (it is within the standard deviation range). The reason is that this blend even at Tin 423 K exhibits significant HR during the early reactions (Fig.6) due to the high octane reactivity and n-heptane content. T65RFn5 which has a RON≈105, does not auto ignite at 323K intake temperature and within the CR range of the experimental apparatus. Therefore there is not any data available for the comparison. T50RFm30 (RON≈84) has a significantly higher ϕ-sensitivity at the higher Tin. There are two reasons for this effect: first is the high boiling point of toluene (table 3) that prevents it to get completely vaporized at Tin 323 K. Therefore, some toluene molecules do not engage in the early reactions. This, results in a lower effect toluene on the cool flame and the main combustion detainment. The second reason is a relatively low early reaction HR that cannot provide the required heating before the start of the main combustion event and engaging toluene molecules in the combustion reactions.

The other blend component which affects the ignition delay and fuel ϕ-sensitivity is ethanol. As previously explained in section 3.1 ethanol has quenching effect on LTHR and reacts as a radical sink(49,73,79–82). This study shows that ethanol has a stronger quenching effect on LTHR compared to toluene which is also reported in in the literatures (25,76,81,82). Figure. 3 displays the clear link between HoV of the blends and the percentage of released heat during the early reactions. The responsible component for high HoV in these blends is ethanol. Besides the

scavenging effect of ethanol on LTHR, the high HoV of ethanol blends together with the low stoichiometric ratio of it, provides the charge cooling effect at Tin 323 K which is lower than the boiling point of Ethanol (table .3). The low stoichiometric ratio of ethanol is 9:1 and it means that the required fuel mass for a fuel containing ethanol is higher than other blends of this study and results in higher charge cooling effect. Both the high HoV and the high fuel mass increase the possibility of thermal stratification inside the combustion chamber.

4 Conclusion

The ϕ-sensitivity property of a fuel has a key role in auto-ignition and therefore combustion behaviour of different fuels. ϕ-sensitivity is a temperature-pressure sensitive property and, at different intake temperatures and pressures the ϕ-sensitivity of a fuel might defer. In this study 11 different blends (TERFs) in three research octane number (RON) groups of 63, 84, and 105, together with neat ethanol are evaluated using a cooperative fuel research engine.

- This study suggests that the Lund ϕ-sensitivity number can provide knowledge about ϕ-sensitivity of diverse fuels at different intake temperatures. ϕ-sensitivity evaluation of fuels at different intake temperature, facilitate the optimum fuel tailoring for modern combustion concepts or a combination of different technologies. This study specifically suggests that :
- At 323K intake temperature (≈ The intake temperature of RON test method) the coexistence of oxygenated and aromatic hydrocarbons together with paraffin maximizes the Lund ϕ-sensitivity of the blends, while at the intake temperature of 423 K (≈ The intake temperature of MON test method) the unoxygenated octane sensitive blends (Toluene reference fuel: TRFs) have the highest Lund ϕ-sensitivity number in each research octane number (RON) group.
- Toluene has quenching effect both on cool flam and main combustion and therefore increases the ignition delay of the blends and therefore increases the ϕ-sensitivity of them. At 323K intake temperature, the charge cooling effect of ethanol compound of the blend,

increases the fuel capability of adapting a higher CR that results in a higher in-cylinder pressure and therefore, increases the fuel ϕ-sensitivity.

- The results show, the intake temperature variation does not affect the Lund ϕ-sensitivity numbers of different blends with the same research octane number (RON) in an analogous way. Therefore evaluation of ϕ-sensitivity of different fuels together with the RON and MON measurements is needed to tailor load adaptive fuels for different combustion concepts.
- This study confirms the effect of early reactions heat releases (ERHR) on the ϕ-sensitivity of different fuels independent of intake temperatures and consequently, on the Lund ϕ-sensitivity number.

5 CRediT authorship contribution statement

Nika Alemahdi: Investigation, Formal analysis, Software, Visualization, Writing - original draft. **Antonia García** Supervision, review & editing. **Martin Tuner:** Supervision, review & editing.

6 Declaration of Competing Interest

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

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