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Investigation of Lubricant Oil influence on Ignition of Gasoline-like Fuels by a Detailed Reaction Mechanism

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Abstract

A reaction mechanism, suitable for simulating the lubricant oil influence on the combustion process of gasoline-like fuels, is developed. The proposed work is motivated by evidences reported in the literature highlighting that lubricant oil droplets can be the most likely inducer of pre-ignition phenomena, as well as the fact that lubricant oil can represent the main source of very fine soot particles emitted from the engine. In other words, the mixture of fuel and lubricant oil, through complex physical and chemical reactions involving long hydrocarbons chains, can auto-ignite before the spark timing or, if not the case, can become one of the major candidates in the generation of soot precursors. Therefore, developing a reliable reaction mechanism able to simulate the oil-fuel mixture behavior and contribution in terms of pre-ignition and soot formation is fundamental for predicting the onset of knocking phenomena and particle size distributions of soot emissions. In this study, surrogate components reproducing the lubricant oil propensity to ignition were first identified. A detailed reaction mechanism was then developed and validated starting from existing mechanisms proposed for the single species. In particular, alkanes ranging from C_{16} to C_{18} were selected as oil surrogates. Zero-dimensional numerical simulations were conducted in order to validate the proposed mechanism versus literature experimental data aimed at reproducing the effect of commercial lubricants on ignition propensity of gasoline-like fuels. From this analysis it was possible to define a suitable composition of the surrogate mixture proposed to model lubricant oil effects.

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Keywords: Lubricant Oil Oxidation, Low-Speed Pre-Ignition, Super-knock, GDI Engines, Soot

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1. Introduction

Among internal combustion engine specialists, a recent widespread opinion is that gasoline engines can count on better life perspectives, in comparison with diesel engines. Namely, they have a greater potential for further improve their efficiency and meet upcoming stringent emission regulations [1–4]. In fact, recent research trends have pushed the introduction as common practice of high boost coupled with downsizing and direct injection strategies, offering the potential to enhance power density and reduce fuel consumption in gasoline Spark-Ignition (SI) engines [5]. Nevertheless, the development of such technologies is far from being considered completely mature and is challenging for engine designers, especially concerning the direct injection technology at low-speed and high-load operating regimes [6–9]. The occurrence of abnormal combustion modes represents the main obstacle, which prevents current engines from achieving optimized combustion phasing and higher compression ratios [10–12].

The main, well-known, undesired phenomena is knock [13], and, recently, a new form of abnormal combustion, which is often observed at low engine speed, has become of great concern [14,15]. As summarized by Wang et al. [1], it has been termed in many different ways, but the most widespread expressions are super-knock and Low-Speed Pre-Ignition (LSPI). Such a phenomenon is significantly different from conventional knock, in which the end-gas autoignites ahead the spark-triggered propagating flame, because the pressure and temperature of the unburned mixture are excessively increased by the compression effect due to the thermal expansion of the burned zone (which can overlap that of the moving piston). Rapid Compression Machine (RCM) experiments [16–20] demonstrated that the mechanism of super-knock consists of hotspot-induced deflagration-to-detonation transition, followed by highpressure oscillation. In other words, a local "hot-spot" in the combustion chamber can be the cause of a surface preignition before top dead centre. A pre-ignition-triggered deflagration (flame propagating at subsonic speed) starts the combustion. Then, the spark ignition occurs, and a second flame front may propagate if the spark ignition is in an unburned zone. The rapid expansion of the burned gas quickly compresses the unburned mixture to higher temperature and pressure. Finally, a second hot-spot in the end gas induces the detonation (supersonic propagating waves) of the unburned mixture at high temperature and high pressure.

A single super-knock event is sufficient to severely and instantaneously damage an engine due to the extremely high peak pressure and the associated pressure oscillations involved [1,7,15]. Furthermore, super-knock events appear randomly and show little direct relationship to engine control parameters [21], so that the use of common knock suppression methods, such as retarding spark timing, cooling the intake charge, and enhancing heat transfer, are not effective ways for avoiding super-knock [1,9,22]. Therefore, super-knock is at present the major obstacle for further improving the boost level of turbo-charged SI engines.

Many studies have been conducted to investigate super-knock [6,11–13,23,24], and it is generally accepted that the causes of pre-ignition may relate to lubricant-fuel interactions within the combustion chamber, more than to solely fuel properties. The most popular opinion is that autoignition occurs at hot-spots containing some lubricant oil, which could initiate a propagating flame because long-chain lubricant oil components have shorter ignition delays and oil additives can have a catalytic enhancement effect [8]. In fact, studies based on combustion visualization and numerical simulation have highlighted that the ignition delay of homogeneous stoichiometric air-fuel mixtures is too high for a spontaneous pre-ignition event before spark ignition [8,25]. Thus, only local and stochastic heterogeneities within the air-fuel mixture or external contaminants like oil or deposits, could explain higher local reactivities for premature auto-ignition. Moreover, the stochastic nature of oil intrusion into the combustion chamber represents additional evidence [1,26,27].

Oil droplets are not the only cause of pre-ignition. Floating solid particles in the combustion chamber also have a close connection with pre-ignition, in the sense that higher in-cylinder soot emissions increase the frequency occurrence of pre-ignition and super-knock cycles in gasoline direct injection engines [8,22,28-31]. As a product of incomplete combustion, particles can stay in the combustion chamber and serve as high temperature spots able to ignite the mixture. It has been demonstrated that a clean combustion chamber without carbon deposits clearly shifts to higher values the pressure level at which pre-ignition occurs [12]. Similarly, increasing spray-wall interaction strongly increases the probability of pre-ignition events [32]. It must be considered that oil contamination within the combustion chamber can represent a non-negligible source for soot particle generation as well, as highlighted by recent literature findings [33,34]. The presence of hydrocarbons with longer chains or more complex structures during the combustion event can enhance soot precursor formation in the reaction zone [35,36], including C₂ species, such as the ethyl radical (C₂H₅) and acetylene (C₂H₂), which are the most abundant gaseous hydrocarbon species in regions where soot is formed [37].

For all the above-mentioned reasons, it is crucial to study the reaction paths involved in the combustion of lubricant oils. In the literature, there is an evident lack of dedicated research on autoignition delay times of lubricant oils. This is instead essential for developing reliable reaction mechanisms useful at predicting the onset of super-knocking phenomena, as well as soot formation in SI engines [37–39]. In order to provide some tentative guidance, Kalghatgi et al. [8] analysed the influence of the addition of n-heptane to iso-octane. It was observed that the ignition delay values were still too high for autoignition to occur at the measured pre-ignition pressures and estimated temperatures. The conclusion was that to satisfy ignition delay time and critical initiating flame size requirements, the oil would have to be somewhat more reactive than n-heptane.

This study defines a way for taking into account the presence of lubricant oil in the combustion process involving gasoline-like fuels. In particular, a detailed reaction mechanism, containing selected surrogate species for lubricant oil, is developed ad validated by means the comparison between 0D numerical simulations and experimental results reported in the literature.

2. Materials and methods

In the present work, an approach based both on the analysis of exiting experimental data and the accomplishment of numerical simulations was adopted. Namely, the analysis of experimental evidences available in the literature allowed the definition of hydrocarbon species suitable for reproducing oil's chemical properties in terms of ignition propensity. Once defined the surrogate species, a detailed reaction mechanism was developed and validated. In order to test the developed mechanism, 0D numerical simulations were conducted using the CHEMKIN PRO code. The closed homogeneous batch reactor model was employed for solving the time-dependent balance equations for the total mass, the gas-phase species and the energy [40]. The simulations aimed at reproducing experimental measurements available in the literature, carried out to quantify the effect of commercial lubricants on the ignition delay time of iso-octane, selected as representative surrogate for commercial gasoline. The comparison between numerical results and experiments, allowed eventually the definition of a possible composition for a surrogate mixture of hydrocarbons that is suitable for modelling the influence of lubricant oil.

2.1. Definition of the surrogate components for modeling engine lubricant oil

All Chemical analyses indicate that base oils are mainly composed of C_{15} – C_{34} normal alkanes (n-paraffins) combined with other iso-alkanes and cycloalkanes [41,42]. Considering that it is known that alkanes larger than C_{14} exhibit nearly identical fuel/air gas-phase ignition delay times across a range of operating temperatures [43,44], surrogate components comprising linear or low-branched alkanes having the number of carbon atoms ranging from 15 to 20 can be considered good candidates at representing the ignition propensity and ignition kinetics of lubricant oils for engine applications.

In order to quantify the effects of oil addition on the ignition propensity of gasoline, Kuti et al. [41] carried out a fundamental experiment involving an Ignition Quality Tester (IQT), in which lubricant base oils, such as SN100 (Group I) and HC4 and HC6 (Group III), as well as a fully formulated lubricant (SAE20W50), were mixed, in the relative amounts of 1% and 10% by volume, with iso-octane (iC₈H₁₈). Successively, with the aim to provide some guidance about the choice of oil surrogate components, the same experiment was repeated by replacing the oils with $nC_{16}H_{34}$, $nC_{17}H_{36}$ and $nC_{18}H_{38}$. These species were mixed with iso-octane in the same proportions used for the oils. A comparison between the recorded trends is reported in Figure 1.

It is noteworthy that the addition of 1% by volume of base oils or fully formulated lubricants into 99 % by volume of iso-octane resulted in ignition delays being averagely shortened by 15%, compared to 100% iso-octane, as shown in Figure 1 (a) and (b), respectively. Further increase in their amount to 10% in the mixtures reduced the average total ignition delay time by up to 54%. Base oils and fully formulated lubricant highlighted a quite similar behaviour, suggesting that it is the hydrocarbon fraction that contributes primarily to enhance the reactivity, and not the inorganic or organometallic additives. This result implies that it is reasonable to neglect the additives' effect in the development of a reaction mechanism for lubricant oil. The experiments in which $nC_{16}H_{34}$, $nC_{17}H_{36}$ and $nC_{18}H_{38}$ were added to pure iso-octane showed trends that appeared to be really close to those obtained for oil lubricants. This result suggests that $C_{16}-C_{18}$ hydrocarbons are adequate surrogates for capturing the ignition characteristics of commercial engine lubricants and thus a reaction mechanism for iso-octane, comprising such hydrocarbon species, is proposed in the

present work. In particular, the model employed in the present works comprises n-C₁₆H₃₄ and C₁₈H₃₈₋₂ as surrogate hydrocarbon species for lubricant oil, for which accurate mechanisms are available in the literature.

2.2. Mechanism development and validation

The chemical kinetic model presented in this work aims to capture lubricant oil influence on pre-ignition occurrence in gasoline engines. It is developed starting from existing reaction mechanisms available in the literature, already validated for the species discussed in the previous section. In particular, the reduced Primary Reference Fuel (PRF) mechanism proposed by Wang et al. [45] based on 73 species and 296 reactions, was used to reproduce the behavior of iso-octane. The species and reactions involved in the mechanism developed by Sarathy et al. [44] for 2-methylalkanes up to C_{20} and n-alkanes up to C_{16} , containing approximately 7200 species and 31400 reactions, were added to the PRF mechanism. It must be highlighted that, unfortunately, to the authors' knowledge, no reduced mechanisms for alkanes up to C_{20} are available in the literature. Therefore, a detailed mechanism was used for those species and the resulting mechanism consists of 7182 species and 31721 reactions.

A thorough analysis of reactions and species involved in the two original mechanisms was performed in order to ensure that the merging process does not affect the agreement with the experimental data against which the starting mechanisms were originally validated. Therefore, the resulting reaction mechanism was validated against the literature data already used for validating the mechanisms by Wang et al. [45] for iso-octane and n-heptane and by Sarathy et al. [44] for 2-methylhexane, respectively.



Figure 1. Variations of ignition delay with temperature for iso-octane and lubricant base oil mixtures, iso-octane and SAE20W50 lubricant mixtures and iso-octane and C₁₆–C₁₈ n-alkanes adapted from Kuti et al. [41].



Figure 2. Simulations using the present mechanism (solid lines) and comparisons with literature experimental data (marks) and mechanisms (dashed lines). (a) shock-tube experiments by Fieweger et al. [46] using iso-octane and n-heptane and simulations by the PRF mechanism of Wang et al. [45]; (b) Galway RCM experiments by Silke et al. [47] using 2-methylhexane and simulations by the mechanism of Sarathy et al. [44].

Figure 2 (a) shows the results in which 0D homogeneous constant-volume reactor simulations (solid lines) were performed to reproduce the shock-tube experiments carried out by Fieweger et al. [46] (marks) in the case of pure iso-octane and n-heptane. A pressure of 40 bar and stochiometric conditions were considered in this case, as it was in the experiments. Figure 2 (b) shows the results of 0D RCM kinetic modeling of the tests performed by Silke et al. [47], concerning 2-methylhexane. In the last case, the closed homogeneous batch reactor model was used to simulate the RCM experiments in a temperature range 600-900 K, considering a pressure of 15 atm and an equivalence ratio equal to 1. The onset of ignition was estimated as the point of maximum temperature rise (max dT/dt). In Figure 2 (a) and (b) the dashed lines refer to the numerical predictions obtained by using the reaction mechanisms developed by Wang et al. [45] and Sarathy et al. [44], respectively.

The results obtained by means of the proposed reaction mechanism are in good agreement with the experimental data and are close to those obtained by the numerical simulations employing the two original mechanisms. This result confirms that the merging process did not affect the ability of capturing the ignition delay trends of the pure species considered for the validation.

3. Numerical simulations

Using the proposed detailed reaction mechanism including lubricant oil surrogate components, numerical simulations were performed to test its capability of predicting the lubricant oil influence on ignition propensity of gasoline-like fuels. In order to pursue this goal, 0D simulations aimed at reproducing the experimental IQT tests, carried out by Kuti et al. [41], were performed. The same procedure used for estimating the ignition delay in the experiments was used in the numerical simulations, namely it was based on the time of maximum pressure rate rise (max dp/dt).

3.1. Operating conditions

In the numerical simulations, the surrogate hydrocarbon species (n-C₁₆H₃₄ and C₁₈H₃₈₋₂) selected for modeling lubricant oil were mixed with iso-octane, in the amounts of 1 and 10% by volume. The ignition delay of the mixtures was calculated considering a closed homogeneous batch reactor model. In the experiments, an ambient pressure of 15 bar was maintained in an IQT with a volume of 210 cm³. However, as the fuel was injected into the chamber vaporization took place, and the mixture temperature decreased accordingly. As a consequence, the chamber pressure decreased of about 1.5 bar [41]. Therefore, in order to reproduce the experimental conditions reached at the end of the injection process, a pressure of 13.5 bar was considered in the simulations. The fuel mass injected was kept constant and equal to 83 mg/inj. The initial temperature was varied from 650 to 900 K and the initial mass of air in the chamber changed accordingly. As a result, the global equivalence ratio, Φ , varied from 0.82 to 1.14, as the temperature increased. For clarity, the operating conditions used in the numerical simulations are summarized in Figure 3. It must be highlighted that the different hydrocarbon mixture compositions are characterized by different stoichiometric airto-fuel ratios. However, the relative amount of the species added to iso-octane was small, so that the variations of Φ with temperature for the seven considered cases coincide within plotting accuracy, as shown in Figure 3.



Figure 3. Operating conditions used in the simulations aimed at reproducing the experiments by Kuti et al. [41].



Figure 4. Percentage reduction in ignition delay measured by Kuti et al. [41] due to an addition to iso-octane of 1% (blue marks) and 10% by volume (red marks) of base oils (a) and fully formulated lubricant oil (b). Numerical simulations with n-C₁₆H₃₄ and C₁₈H₃₈₋₂ (dashed lines). Simulations with a binary surrogate mixture (50% n-C₁₆H₃₄ and 50% C₁₈H₃₈₋₂, by volume) added to iso-octane (solid lines).

3.2. Numerical results

Numerical simulations were conducted to assess the accuracy of the present mechanism in predicting the percentage reduction of the ignition delay that gasoline experiences in the presence of lubricant oil contaminants. Figure 4 provides a comparison between the experimentally-measured (marks) and the numerically-predicted (lines) data about the reduction of ignition delay caused by an addition of 1 or 10% by volume of lubricant oil to iso-octane. The marks in Figure 4 (a) represent the average reduction measured for the different base oils employed in the experiments and previously described (see Figure 1 (a)). The experiments reported in Figure 4 (b) are related to the fully formulated lubricant oil SAE20W50 (previously shown in Figure 1 (b)).

Figure 4 shows that the averaged ignition delay reduction derived from the experiments by Kuti et al. [41] is between the values obtained by the two numerical simulations using $n-C_{16}H_{34}$ and $C_{18}H_{38-2}$ (dashed lines). This result was obtained for both the two considered volume fractions and for both base oils and the commercial lubricant oil, confirming the effectiveness of the developed mechanism in reproducing the lubricant oil effect on ignition propensity of iso-octane.

Only at low temperatures a discrepancy appears in the case of 1% volume fraction for the fully formulated lubricant oil, but one can notice that generally, the uncertainty of the experimental measurements increases at low temperatures, especially for temperatures lower than 750 K, as it is possible to infer from Figure 1 and from the error bars reported in Figure 4. Therefore, it is difficult to ascribe the reason of such a discrepancy only to poor accuracy of the reaction mechanism in the low-temperature range. In addition, for all other cases the agreement can be considered more than satisfactory at the lower temperatures.

On the basis of these results, it was possible to determine a surrogate hydrocarbon mixture that could averagely reproduce the reactivity of lubricant oils. Namely, it was concluded that a surrogate binary mixture composed of n- $C_{16}H_{34}$ and $C_{18}H_{38-2}$, taken in the same proportions, can reproduce the higher reactivity of lubricant-fuel mixtures in comparison to that of pure iso-octane with good accuracy. Figure 4 also shows the results obtained from the simulations in which this surrogate mixture was mixed with pure iso-octane (red and blue solid lines). Such a mixture shows an intermediate behavior with respect to $n-C_{16}H_{34}$ and $C_{18}H_{38-2}$, in terms of ignition delay, and therefore can be reasonably assumed as a surrogate mixture capable of reproducing the effects of commercial lubricant oils on gasoline's ignitability.

4. Conclusions

The present work helps to define a way for taking into account lubricant oil presence within an engine's combustion chamber, in order to simulate its contribution to pre-ignition and soot formation processes. On the basis of experimental results available in the literature, hydrocarbons species suitable for reproducing oil's chemical reactivity were first identified. Specifically, it was found that C_{16} – C_{18} hydrocarbons can be considered adequate surrogates that

are able to capture the ignition characteristics of commercial lubricant oils. Then, a detailed reaction mechanism containing the selected surrogate species was developed starting from existing mechanisms. The reduced PRF mechanism proposed by Wang et al. [45] was coupled with the mechanism developed by Sarathy et al. [44] for 2-methyl-alkanes up to C_{20} and n-alkanes up to C_{16} . The resulting mechanism consisted of 7182 species and 31721 reactions. It was validated against the same literature data used for validating the two starting mechanisms. It was found that the merging process did not affect the ability of capturing ignition delay trends of the species considered for the validation. The results obtained by means of the proposed reaction mechanism were very close to those obtained by numerical simulations employing the two original mechanisms.

Once the surrogate species for lubricant oil were defined and the reaction mechanism validated, 0D numerical simulations were conducted in order to reproduce experimental measurements available in the literature, aimed at quantifying the effect of lubricant oils on the ignition delay of iso-octane (selected as surrogate for commercial gasoline). The simulated system was a closed homogeneous batch reactor in which n-C₁₆H₃₄ and C₁₈H₃₈₋₂ were mixed with iso-octane, with 1 or 10% by volume. The results showed that the percentage reduction of the ignition delay measured in the experiments was between the values obtained by the numerical simulations using n-C₁₆H₃₄ and C₁₈H₃₈₋₂. The results demonstrate the accuracy of the developed mechanism in reproducing lubricant oil effect on the ignition propensity of gasoline-like fuels.

Furthermore, by means of the numerical analysis, it was possible to determine a surrogate hydrocarbon mixture that could averagely reproduce the reactivity of both base oils and a fully formulated lubricant oil. In particular, it was observed that a mixture of $nC_{16}H_{34}$ and $C_{18}H_{38-2}$, in the same proportions, can emulate the chemical characteristics of a commercial lubricant oil in a satisfactory way. Future work will arrive at reducing the proposed mechanism to make it suitable for engine simulations.

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