Dynamics analysis of a jet fuel surrogate and development of a skeletal mechanism for CFD applications

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ABSTRACT

The autoignition dynamics of a three component surrogate jet fuel (66.2% n-dodecane, 15.8% n-proplylbenzene, 18.0% 1,3,5,trimethylcyclohexane) suitable for usage as Jet A-1 and RP-3 aviation fuels are analyzed, using the detailed mechanism of Liu et al. (Liu et al. 2019). The conditions considered are relevant to the operation of gas turbines and the analysis is performed using mathematical tools of the computational singular perturbation (CSP) method. The key chemical pathways and species are identified in the analysis of a homogeneous adiabatic and constant pressure ignition system for a wide range of initial conditions. In particular, the key role of hydrogen and CO-related chemistry is highlighted, with an increasing importance as the initial temperature increases. The $C_2H_4 \rightarrow C_2H_3 \rightarrow CH_2CHO$ pathway is also identified to play a secondary but non-negligible role with an importance increasing with initial temperature, favoring the system's explosive dynamics and, thus, promoting ignition. Finally, C_2H_4 is identified to be a species with a key (secondary) role to the system's explosive dynamics but its role is replaced by C_3H_6 and eventually by O_2, as the initial temperature increases. In the second part of the current work, a 58-species skeletal mechanism is generated using a previously developed algorithmic process based on CSP. The developed skeletal was tested in a wide range of initial conditions, including both ignition delay time

and laminar flame speed calculations. For the conditions that were of interest in the current work, the skeletal approximated the detailed mechanism with very small error. The 58-species skeletal is shown to be ideal for use in CFD applications not only because of its small size but also because of its sufficiently slow associated fast timescale.

INTRODUCTION

Jet aviation fuels, such as Jet A/A1 and JP-8, are typically comprised of hundreds of chemical components. Therefore, the approach to use surrogate fuels (which typically consist of a handful of chemical components) for modelling purposes have been widely accepted as these fuels are able to emulate closely the combustive characteristics of their parent fuels, hence, easing significantly the modelling process. The proper modelling of combustion phenomena on the basis of such surrogates, requires the development of chemical kinetic mechanisms, which, typically consist of hundreds or thousands of hydrocarbon species.

The development of detailed chemical kinetic mechanisms for conventional jet fuel mixtures has mainly grown the last two decades. Li et al. (Li et al. 2001) produced a detailed mechanism of 36 species and 174 reactions for JP-10, which was validated against shock tube data in the range of $\varphi = 0.5$ and 2.0, $p(0) = 1$ and 100 bar, $T(0) = 1000 - 2500$ K. Violi et al. (Violi et al. 2002) developed a semi-detailed chemical kinetic model (72 species and 256 reactions) and proposed a six component surrogate to represent JP-8. Dagaut et al. (Dagaut et al. 2006) reported the development of a detailed kinetic reaction mechanism (209 species and 1673 reactions) and a three component surrogate for Jet A-1, which was tested in PSR and premixed flame simulations in the range of $T(0) = 900 - 1300$ K, $p(0) = 1$ - 40 atm and $T(0) = 800$ K, $p(0) = 1$ atm, $\phi = 1.7$, respectively. Gokulakrishnan et al. (Gokulakrishnan et al. 2007) developed a detailed mechanism and proposed a four component (n-decane, n-propylcyclohexane, n-propylbenzene, and decene) surrogate to represent Jet A. The mechanism was validated against shock tube ignition delay time measurements, jet-stirred reactor reactivity profiles, and plug-flow reactor species time–history profiles. Strelkova et al. (Strelkova et al. 2008) developed a detailed mechanism of 71 species and 417 reactions and a reduced one (24 species and 38 reactions), both for Jet A fuel, and proposed a three component surrogate fuel (72.7% n-decane, 9.1% n-hexane, 18.2% benzene). Both mechanisms were validated against variable experimental data found in the literature in the range of $p(0) = 1, 10, 100$ atm, $T(0) = 1000 - 1800$ K, $\varphi = 0.5, 1, 2$. Honnet et al. (Honnet et al. 2009) developed a detailed chemical-kinetic mechanism (122 species and 900 reactions) to describe the combustion of the Aachen surrogate (n-decane 80% and 1,2,4-trimethylbenzene 20%) as a surrogate of kerosene and tested it against critical conditions of autoignition and soot volume fraction. Dooley et al. (Dooley et al. 2010) developed a detailed chemical kinetic mechanism (1599 species amongst 6633 reactions) for a three component (n-decane, iso-octane and toluene) Jet A POSF 4658 surrogate and validated it against variable pressure flow reactor simulations ($T(0) = 500 - 1000$ K, $p = 12.5$ atm), counterflow flames ($p = 1$ atm, $T_f = 500$ K, $T_o = 298$ K) and autoignition ($T(0) = 625 - 1250$ K, $p(0) = 17 - 23$ atm). Malewicki et al. (Malewicki et al. 2013) generated a detailed mechanism of 2080 species and 8310 reactions for Jet A POSF 4658 fuel, introducing also a four component surrogate fuel (n-dodecane, iso-octane, n-propylbenzene, 1,3,5- trimethylbenzene). The mechanism was validated against shock tube data, in the range of $\varphi = 0.46, 1.86$, $T(0) = 879 - 1733$ K, $p(0) = 16 - 27$ atm for the POSF 4658 and in the range of $\varphi = 0.77, 1.85$, $T(0) = 879 - 1733$ K, $p(0) = 16 - 27$ atm for the surrogate. Gao et al. (Gao et al. 2015) developed a detailed mechanism of 691 species and 15518 reactions for JP-10 fuel. The mechanism was validated against shock tube experimental data in the range of $\varphi = 0.14$, $T(0) = 1000 - 2000$ K, $p(0) = 1$,1.7, 7 & 20 atm. Yu and Gou (Yu and Gou 2018) produced a detailed mechanism (1880 species and 6586 reactions) for the purpose of using it for surrogate fuel models of S-8, Jet-A, or RP-3. Using path flux analysis they generated two skeletal mechanisms one for high temperatures (203 species and 1131 reactions) and another for low temperatures (720 species and 3085 reactions). Both the detailed and the simplified mechanisms were validated against ignition delay times ($T(0) = 670 - 1250$ K, $\varphi = 1.0$, $p(0) = 10 - 20$ atm) and laminar burning velocities ($T_u = 400$ K, $\varphi = 0.7 - 1.4$, $p(0) = 1 - 3$ atm). Finally, Liu et al. (Liu et al. 2019) developed a detailed mechanism of 401 species and 2838 reactions for a three component surrogate for Jet A1 and RP-3 (66.2% n-dodecane, 15.8% n-proplylbenzene, 18.0% 1,3,5,trimethylcyclohexane), which was validated in the range of $\varphi = 0.6 - 2.0$, $T(0) = 500 - 1700$ K, $p(0) = 1 - 16$ atm, against various experimental configurations: shock tube, laminar burner and jet stirred reactor.

Detailed reaction mechanisms like those previously described can be readily employed in zero-dimensional (0D) simulations, however, their size can be prohibitive even for 1D simulations, let alone 2D or 3D simulations which are of more practical interest. For this purpose, various mechanism simplification and reduction techniques are employed, such as the computational singular perturbation and the sensitivity analysis. Through these simplification techniques, the size of the detailed mechanism is systematically reduced by removing species and reactions with little impact to a particular feature, which can be as trivial as the ignition delay time or more sophisticated like the system's dynamics. Depending on the outcome of the simplification process (i.e., the size of the generated mechanism), the produced skeletal can be used as is in computational fluid dynamics (CFD) simulations or an additional reduction step may have to follow.

Therefore, the development of simplified and reduced reaction mechanisms for aviation fuels has flourished, especially the last 10 years, with the tremendous advances in computational power, which have enabled the simulation of reacting flow phenomena with multi-step reaction schemes. Munzar et al. (Munzar et al. 2013) reported the development of a skeletal mechanism (173 species and 1197 reactions) using a species sensitivity reduction method, referred to as the Alternate Species Elimination (ASE) method, and validated it against the laminar flame speed of various jet fuel surrogate components (n-decane, methylcyclohexane and toluene). Tosatto et al. (Tosatto et al. 2013) used directed relation graph (DRG) techniques to develop various skeletal mechanisms (82-92 species) which were validated for a two component JP8 surrogate against 2D coflow flames. Tay et al. (Tay et al. 2016) developed a skeletal kerosene-diesel reaction mechanism (123 species and 586 reactions) with embedded soot chemistry for diesel engine simulations and validated against ignition delay time, heat release rate, flame lift-off lengths and in-cylinder soot and pressure ($p(0) = 20, 60$ and 67 atm, $\varphi = 0.25 - 2.00$, $T(0) = 700 - 1400$ K). Yu et al. (Yu et al. 2018) developed a 74-species (189 reactions) skeletal mechanism optimised on the basis of a five component (n-dodecane/iso-octane/iso-cetane/decalin/toluene) jet fuel surrogate of Jet A POSF-4658 and tested it in 0D and 3D simulations. Fang et al. (Fang et al. 2018) developed a 3-component jet fuel surrogate (methylcyclohexane, n-dodecane and m-xylene) mechanism (110 species and 352 reactions) through semi-global sub-mechanism construction and mechanism reduction and validated it against ignition delays in shock tube ($T(0) = 650-1300$ K, $\varphi = 0.5-1.5$, $p(0) = 5-80$ atm), laminar flame speeds ($T_u = 400$ and 470 K, $\varphi = 0.7-1.4$, $p = 1-3$ atm) and species concentration profiles in shock tube ($T(0) = 900-1600$ K, $\varphi = 0.46-1.85$, $p(0) = 16-25$ atm) and plug flow reactor ($T(0) = 500-1100$ K, $\varphi = 1.0$, $p(0) = 12.5$ atm). Yi et al. (Yi et al. 2019) produced a skeletal kerosene chemical kinetic mechanism (231 species and 5591 reactions) aiming at the properties of the RP-3 fuel through a 4-component surrogate (toluene/trans-decalin/n-decane/iso-cetane) and assessed it for predictions of assessed for predictions of the distillation curve and physicochemical properties as well as combustion properties of ignition delay times ($T(0) = 1000-1500$ K, $\varphi = 0.5-1.5$, $p(0) = 1-10$ atm) and laminar flame speeds ($T_u = 403$ K and 470 K, $\varphi = 0.65-1.4$, $p = 1$ atm).

Although the aforementioned simplified mechanisms offer a solid basis for further research activity, most of them are still quite large in size to be employed in computationally affordable multidimensional CFD simulations. The mechanisms of Yu et al. (Yu et al. 2018) and Tosatto et al. (Tosatto et al. 2013) are probably the only ones which can be employed in multidimensional CFD simulations but none of them was optimised against Jet A1 (the target fuels were Jet A POSF-4658 and JP8, respectively) and their size can still pose a significant challenge in direct numerical simulations (DNS). In addition, to the best of the authors' knowledge, there has been no systematic computational analysis of the key chemical pathways in the combustion of conventional jet fuel surrogates. This is particularly important in the design of modern efficient and environmental friendly propulsion systems for the aviation sector. Sporadic reports exist though in the literature, where computational analysis was performed with reaction flux analysis (Strelkova et al. 2008; Liu et al. 2019; Banerjee et al. 2016; Wang et al. 2018; Kukkadapu et al. 2019; Dagaut 2006) and/or brute force sensitivity analysis (Liu et al. 2019; Banerjee et al. 2016; Wang et al. 2018; Gokulakrishnan et al. 2007; Naor et al. 2018; Zhong and Peng 2019; Mawid and Sekar 2006; Raza et al. 2019; Humer et al. 2007; Seshadri et al. 2011). However, none of these was systematic and typically one set of conditions was used. Furthermore, except for the works of (Liu et al. 2019; Zhong and Peng 2019; Mawid and Sekar 2006; Dagaut 2006), the rest of them employed in the respective (reaction flux/sensitivity) analysis single-component surrogates such as n-decane (e.g., (Humer et al. 2007; Gokulakrishnan et al. 2007; Raza et al. 2019)) and n-dodecane (e.g., (Humer et al. 2007; Banerjee et al. 2016; Naor et al. 2018)). Moreover, the analysis was performed in low to intermediate temperatures for all the aforementioned works, except for the works of (Strelkova et al. 2008; Banerjee et al. 2016; Humer et al. 2007) which were indeed conducted on the basis of high temperatures. Most important, both brute force sensitivity analysis and reaction flux analysis provide evidence that is only global in character, thus, missing valuable information and details that can only be picked up by more sophisticated methods such as the computational singular perturbation approach (CSP) (Lam and Goussis 1989). In other words, neither brute force sensitivity analysis nor reaction flux analysis can elucidate the temporal/spatial evolution of the set of reactions and species that have the larger effect on the system's dynamics.

Hence, the objective of the current work is twofold. Firstly, to obtain physical understanding on the key processes that control the combustion of an aviation jet fuel (Jet A1) in view of a surrogate, thus, enabling the design of more efficient engines, and secondly, to develop a skeletal model, sufficiently small for usage in CFD, thus offering the scientific community and the industry the capability to investigate and test efficiently new engine design configurations. The emphasis of this work will be on high temperatures ($1,200\ K < T(0) < 1,800\ K$), fuel lean and medium pressures of $10\ atm < p(0) < 20\ atm$, i.e., conditions relevant to the operation of gas turbines. It is noted that the fuel lean conditions are employed because they can ensure a reduction in the formation of pollutants, namely soot and NOx, compared to the fuel rich types of combustion that are used in more conventional aeropropulsion systems (Lefebvre and Ballal 2010). Both goals, will be accomplished through the employment of advanced mathematical tools and methods, part of the computational singular perturbation approach (CSP) (Lam and Goussis 1989). For the purposes of the current work, the kinetic mechanism of Liu et al. (Liu et al. 2019) will be used with an emphasis on the three component surrogate that was introduced. The aforementioned mechanism was selected because of its proven accuracy in a wide range of operational conditions and a variety of configurations.

The structure of the paper is as follows. Firstly, the mathematical tools will be briefly described along with the simplification method, all part of the computational singular perturbation approach. Next, a dynamics analysis of a homogeneous autoigniting system will be performed, in a wide range of initial conditions, where the key processes and variables controlling the ignition of the system will be identified. Afterwards, the simplification process will be presented along with the produced skeletal mechanism and evidence of its validity in a wide range of conditions will be demonstrated.

THE MATHEMATICAL TOOLS

In this work, mathematical tools of the CSP approach are used, an algorithmic method of asymptotic analysis introduced in the late 80s (Lam and Goussis 1989). The proposed method has showcased its success the last 30 years in providing reduced models of increased accuracy and analysing in detail highly complex mathematical models, in a wide range of different fields, such as chemical kinetics (e.g., (Tingas et al. 2018c; Yalamanchi et al. 2020)), reacting flows (e.g., (Manias et al. 2019b; Prager et al. 2011)), atmospheric environment (e.g., (Neophytou et al. 2004; Neophytou et al. 2005)), applied mathematics (e.g., (Goussis and Valorani 2006; Maris and Goussis 2015)), biological modelling (e.g., (Patsatzis and Goussis 2019; Patsatzis et al. 2019)) and pharmacokinetics (e.g., (Patsatzis et al. 2016; Michalaki and Goussis 2018)). In the field of combustion, CSP has been used in the analysis of a range of different applications such as zero-dimensional autoigniting systems (e.g., (Tingas et al. 2015; Khalil et al. 2019)), one-dimensional laminar flames/igniting systems (e.g., (Massias et al. 1999; Song et al. 2018)), two-dimensional turbulent igniting systems (e.g., (Pal et al. 2017)) and three-dimensional turbulent flames (e.g., (Manias et al. 2019c; Manias et al. 2019a)). For a detailed description of the CSP method and the tools used in the current study, the reader is referred to (Lam and Goussis 1994; Hadjinicolaou and Goussis 1998; Valorani et al. 2006b; Valorani et al. 2020).

In the CSP framework, the right-hand-side (rhs) of the system of the species and temperature equations is projected to the CSP vectors. Eventually, the rhs of the aforementioned system is expressed as a sum of the CSP modes. Each CSP mode is characterised by: (i) a timescale (τ_i), which sets the timeframe of action of the respective mode and (ii) an amplitude (f^i) which represents the impact of the related mode to the slow evolution. The CSP modes, which are ordered based on their timescales from fast to slow ($\tau_1 < \tau_2 < \ldots$), can be either explosive or dissipative, depending on the sign of the real part of the respective eigenvalue; explosive modes are associated with positive (real parts of) eigenvalues while dissipative modes are associated with negative (real parts of) eigenvalues. Interested in leading order accuracy, the CSP vectors are approximated by the right and left eigenvectors of the Jacobian of the chemical source term (Diamantis et al. 2015a; Singh et al. 2019). In that case, the timescale of the i-th mode is given by: $\tau_i = \frac{1}{|\lambda_i|}$. When the M fast timescales have been exhausted, the system's slow evolution is governed by the characteristic mode with an associated timescale (τ_i) being among the fastest of the slow ones and an associated amplitude f^i among the largest in magnitude. Usually, the characteristic modes are explosive in nature, i.e., tend to drive the system away from equilibrium (Sarathy et al. 2019; Jaasim et al. 2018). Explosive modes are commonly associated with autoigniting phenomena (Tingas et al. 2018b; Tingas et al. 2018d) and flames (Najm et al. 2010; Manias et al. 2018) but as has been highlighted in the recent works of (Manias et al. 2019c; Manias et al. 2019b) the role of dissipative modes in the system's slow evolution cannot be dismissed as some of them may have more important role than the explosive ones. Therefore, the role of the explosive mode as the system's characteristic one is not straightforward and must be subject to investigation, on the basis of the aforementioned criteria.

The number M of the exhausted modes is typically associated with a timescale gap. However, in the CSP framework, M can also be determined algorithmically (Valorani and Goussis 2001). In the current work, M has been calculated using the definition introduced later in (Valorani et al. 2018).

Interested in identifying the key processes and species that control the evolution in time of the autoigniting phenomena, the following CSP tools are used:

- the timescale participation index (TPI), which identifies the chemical reactions with the largest contribution to each CSP mode's timescale (Goussis and Najm 2006; Li et al. 2019). By identifying the system's characteristic CSP mode and the associated timescale, the TPI is used to identify the reactions with the largest contribution to the characteristic timescale (Tingas et al. 2016a; Tingas et al. 2019). These reactions have the largest effect on the system's slow evolution, i.e., can accelerate/decelerate significantly the system's evolution in the physical time (Tingas et al. 2017; Tingas et al. 2015).

- the CSP Pointer, which identifies the variables (species or temperature) which are mostly associated with the each CSP mode. This tool has been used in model reduction to identify the species for which the quasi-steady state assumption (QSSA) holds (Massias et al. 1999; Valorani et al. 2003; Tingas et al. 2018a). In another application, this tool has been used to identify species that can be used as additives in order to accelerate/decelerate the ignition of a mixture (Manias et al. 2016; Tingas et al. 2016b). In this work, the CSP Pointer will be used to identify the species mostly related to the system's characteristic CSP mode.

For the simplification of the detailed mechanism and the development of the skeletal, the method introduced in (Valorani et al. 2006b; Valorani et al. 2006a; Prager et al. 2009) was utilized, which is based on the CSP framework. Very briefly, the reactions are eliminated on the basis of their contribution to the fast and slow dynamics of a set of species. The contribution of each reaction to the fast and slow components of each species is assessed through the importance index (Goussis and Lam 1992; Prager et al. 2011). The screening of the reactions and the species is performed in an iterative fashion. Initially, the user specifies a starting kernel of target species, typically these being the major products of any combustion process, i.e., CO_2 and H_2O. At the beginning of the first iteration, the importance indices of all reactions are calculated and those having a value larger than a user-specified threshold are kept, while the rest are considered unimportant and neglected. At the end of the first iteration, a new set of species has been identified which includes all the species involved in the reactions with sufficiently large importance indices. The union of these two sets of species forms the new set of target species which is used as the initial kernel at the beginning of the next iteration. The procedure ends when the set of target species does not change between two consecutive iterations.

It is noted that CSP simplification process relies heavily on the system's timescale decomposition to fast and slow modes, as those are determined by M. Therefore, in cases where there is no clear timescale gap, the algorithmic determination of M affects (by properly adjusting the CSP absolute and relative tolerances (Valorani et al. 2018)), as it will also be shown next, the accuracy and size of the produced skeletal mechanisms. The advantage of the CSP simplification process against other methods was showcased in the early work of (Valorani et al. 2006b).

For both the dynamics analysis and the simplification of the detailed mechanism, the CSPTk package (csp 2015) integrated with the TChem package (Safta et al. 2011) for thermokinetic database management was employed.

AUTOIGNITION DYNAMICS OF THE DETAILED MECHANISM

The three component surrogate mixture that was used for detailed dynamics analysis consisted of n-dodecane 66.2%, n-propylbenzene 15.8% and 1,3,5-trimethylcyclohexane 18.0%, in mol. Interested in gas turbine relevant conditions, three cases of $T(0) = 1,200, 1500, 1800$ K were selected with initial pressures of $p(0) = 25$ atm and $\varphi = 0.7$. Although not shown, further studies were also conducted at initial pressures of $p(0) = 10$ and 40 atm, and the results were found to be insensitive to these changes. The zero-dimensional homogeneous adiabatic and constant pressure autoignition of the surrogate mixture was modelled using the TChem package (Safta et al. 2011).

The timescales (dissipative and fast explosive ones) that develop in the three cases under study ($T(0) = 1,200, 1500, 1800$ K) are displayed in Fig. 1 along with the temperature evolution. For convenience, only the faster of the explosive timescales are shown in each case ($\tau_{e,f}$), although some slow explosive timescales do exist in all cases, but since they are much slower than the fast ones, they have negligible effect to the system's slow evolution. It is shown that no timescale gap is evident, indicating no clear split between fast and slow timescales. Hence, the fast explosive timescale resides in the middle of a dense pack of dissipative timescales. If the system generating these features was fully non-linear, the consideration of $\tau_{e,f,}$ would be meaningless. However, the fact that all timescales are constant (among them $\tau_{e,f,}$), except for the very slow ones which have negligible effect on the system's slow evolution, suggests that that the dynamics of the fast explosive mode are quasi-linear. In such a case the fast explosive mode can remain the driving mode, on condition that the related amplitude is dominant. Similar profiles been previously reported in the literature for the dynamics of many hydrocarbons (Tingas et al. 2017; Tingas et al. 2018d; Sarathy et al. 2019; Singh et al. 2019).

The time period during which the fast explosive timescale is present is introduced as the explosive stage of the autoignition process (Diamantis et al. 2015b). The investigation of the system's amplitudes in each case, confirms that the fast explosive mode is always associated with a dominant amplitude, during the explosive stage. It is also highlighted that when using the algorithmic criterion for the determination of M-exhausted modes (orange circle signs in Fig. 1), the fast explosive timescale is always among the fastest of the slow ones, thus, meeting the necessary criterion for being the system's characteristic one. This is a new finding, that has not been previously reported, that reinforces further the fact that the fast explosive mode controls the system's slow evolution.

The explosive timescales (both fast and slow) that develop during the explosive stage of the autoignition of the surrogate fuel at the three different sets of initial conditions are more clearly displayed in Fig. 2. It is shown that $\tau_{e,f}$ develops throughout the explosive stage, while $\tau_{e,s}$ develops only at the end of the explosive stage, a feature common to all cases. In fact, at the end of the explosive stage, $\tau_{e,f}$ meets $\tau_{e,s}$ and then both disappear, as a result of the result of the approach of the two corresponding positive eigenvalues and their disappearance soon after they meet. It is noted that the end of the explosive stage coincides roughly with the ignition delay time (Diamantis et al. 2015b; Tingas et al. 2015).

In order to investigate the reactions that relate mostly to $\tau_{e,f}$ (i.e., the system's characteristic timescale) 5 points across the explosive stage were selected, representative of the whole process: the first one (P_1) being at $t = 0$ s, the second one (P_2) being at $t = 0.25t_{ign}$, the third one (P_3) at $t = 0.5t_{ign}$, the fourth one (P_4) at $t = 0.75t_{ign}$ and the last one (P_5) being at $t = t_{ign}$. All five points are shown in Fig. 2 for all three cases under study and Table 1 displays all the reactions that appear in the discussion next. It is noted that the symbols “f" and “b" that appear in the numbered reactions, stand for forward and backward, respectively. Table 2 includes the larger TPI and CSP Po values at the points that appear in Fig. 2. These are discussed next.

In the first case, i.e., $T(0) = 1,200$ K, initially, hydrogen chemistry plays an important role, favoring the system’s explosive dynamics mainly through the dissociation of H_2O_2 (i.e., reaction 1352f) and the chain branching reaction 1330f. However, a closer investigation of the rest of the important reactions reveals that H-abstraction reactions which lead to n-dodecane isomers (i.e., reactions 2568-2572, $nC_{12}H_{26}$+$HO_2 \rightarrow C_{12}H_{25}$+$H_2O_2$) account for 11.66% of the total contribution to $\tau_{e,f}$ and become the major driver of the explosive dynamics. The identification of HO_2 and H_2O_2 by the CSP Po is fully reasonable considering that both are involved in reactions with the largest TPI values.

Later in the process, hydrogen-related reactions continue to play an important role favoring the system’s explosive dynamics, mainly through reaction 1330f. Notice, that reaction 1341f competes with 1330f, yet, it favors the system’s explosive dynamics for the most part of the explosive stage. This is an unexpected finding, since normally competing pathways will have opposite behaviors in the system’s dynamics. Another key reaction to the system’s dynamics is 1545f, an H-abstraction reaction of C_2H_4, favoring the explosive character of $\tau_{e,f}$. This reaction has been found to play significant role in the ignition dynamics of other hydrocarbons as well such as n-hexane, n-heptane and iso-octane (Tingas et al. 2018d; Singh et al. 2019). Moreover, reaction 1277f is found to favor notably the explosive dynamics, thus accelerating ignition, especially in the middle of the explosive stage (points P_2, P_3, P_4), as has already been suggested in (Liu et al. 2019). The major opposition to the explosive dynamics is produced by the hydrogen related reactions 1350f and 1348f, both chain termination reactions, and the carbon-related chain termination 1517f (recombination of two CH_3 molecules to C_2H_6).

At the end of the explosive stage (point P_5), hydrogen chemistry dominates along with the decomposition of HCO to H and CO, which is in agreement with the dynamics analysis of many hydrocarbons (Jaasim et al. 2018; Singh et al. 2019; Tingas et al. 2018d; Kazakov et al. 2006;

Kooshkbaghi et al. 2015). The CSP Po identifies the temperature as the variable mostly related to the fast explosive mode at the very early stage of the explosive stage (P_2), which indicates that system enters the thermal runway very early, while the chemical runway is significantly suppressed. In addition, C_2H_4 and HO_2 are also identified by the CSP Po, the first one being a reactant in reaction 1545f and the latter being a reactant in 1277f, 1350f, 1411f and 1348f.

At 1,500 K, the contribution of hydrogen-related chemistry to the system's characteristic timescale becomes more pronounced throughout the explosive stage, except for the very beginning of the process, which is still carbon dominated. The chain branching 1330f becomes the major contributor with a contribution gradually increasing from 5% to 25%, favoring the system's explosive dynamics. Unlike the previous case, the chain carrying reactions 1280f and 1282f become important contributors with a joint contribution of 6.5 %, favoring the explosive character of $\tau_{e,f}$. They both relate to the oxidation of the allyl radical, a key intermediate species in the oxidation of n-dodecane, that leads to the formation of CH_2O and CH_2CHO or CH_3CO. Another difference at the initiation of the two cases (1,200 K and 1,500 K) relates to the secondary role of a hydrogen-related reaction; at 1,500 K, 1352f does not play any role but 1341f does. Finally, at 1,500 K, the competing CO formation reactions 1381f and 1386f, contribute fairly to $\tau_{e,f}$, the first one favoring (for being a chain branching) and the latter opposing (for being a chain carrying) the explosive character of $\tau_{e,f}$. At this early stage, the CSP Po identifies two of the three fuel components along with temperature, as the variables mostly related to the fast explosive mode.

Later in the process, the H-abstraction reaction 1545f remains a notable contributor, promoting the system's explosive dynamics, and C_2H_4 being a reactant plays a key role, as it was found to be the case at 1,200 K. Reaction 1341f, continues to play a secondary role, as the second largest hydrogen-related contributor to $\tau_{e,f}$ favoring the explosive dynamics, but the major opposition of hydrogen-related reactions relate to the chain termination 1348f and 1350f, both identified in the 1,200 K case as well, although the relative contribution of 1350f is significantly decreased. On the other hand, the chain carrying reaction 1277f decreases and the chain branching reaction 1559f increases their relative contributions to $\tau_{e,f}$ at 1,500 K. During the interim part of the explosive stage, the CSP Po identifies C_2H_4 (like at 1,200 K), but CH_2O and C_3H_6 are also identified as variables highly related to the fast explosive mode, suggesting that these two species have a measurable effect on the system's slow evolution.

At the end of the explosive stage, $\tau_{e,f}$ is mainly generated by hydrogen and CO related reactions as it was found to be the case at $T(0) = 1,200$ K, although the CO-to-CO_2 reaction 1359f does have a notably larger contribution compared to the $T(0) = 1,200$ K case.

At the highest initial temperature of $T(0) = 1,800$ K, initially, the relative contributions of the hydrogen-related chain branching 1330f and the HCO-to-CO reactions 1386f and 1381f become significantly enhanced compared to the 1,500 K case. The rest of the important contributors to $\tau_{e,f}$ establish the $C_2H_4 \rightarrow C_2H_3 \rightarrow CH_2CHO$ pathway through reactions 1540f and 1559f. The last part of this pathway (i.e., reaction 1559f) competes with 1553b; apparently, 1559f favors the explosive dynamics as a chain branching reaction while 1553b is a chain carrying step. Like in the previous two cases, the CSP Po highlights the important role of C_2H_4 to the system's explosive dynamics.

As it was shown to be the case at 1,500 K, later in the process, the hydrogen chain branching 1330f dominates the explosive dynamics. The remaining important carbon-related reactions maintain the $C_2H_4 \rightarrow C_2H_3 \rightarrow CH_2CHO$ pathway (reactions 1540f/1545f, 1559f), while 1553b competes with 1559f as previously described. Moreover, CO production reactions (1381f, 1733f, 1359f, 1730f, 1385f) become significant contributors to $\tau_{e,f}$, the first three favoring and the latter two opposing the explosive dynamics. The last part of the explosive stage is governed almost exclusively by hydrogen and CO-related reactions, in agreement with the previous two cases. Finally, the CSP Po identifies the temperature as the variable mostly related to the fast explosive mode along with C_3H_6 and O_2.

In summary, as the initial temperature increases:

- The hydrogen and CO-related reactions (mainly 1330f and 1381/1359f, respectively) become more pronounced, i.e., the related reactions become larger contributors to $\tau_{e,f}$.
- The $C_2H_4 \rightarrow C_2H_3 \rightarrow CH_2CHO$ pathway (reactions 1540f/1545f and 1559f) plays a larger secondary role favoring the system's explosive dynamics.

- The thermal runway suppresses the chemical runway, as this is indicated by the larger CSP Po values that temperature reaches for the same t/t_{ign} time instants.
- The secondary role of C_2H_4 is gradually replaced by C_3H_6 and eventually by O_2.

MECHANISM SIMPLIFICATION

The simplification process starts by determining the conditions where the produced simplified mechanism will be validated. Interested in gas turbines' operating conditions, only medium to high initial temperatures were targeted for ignition, i.e., in the range of $1,100K < T(0) < 2,000K$ and $300\ K < T_u < 470\ K$ for laminar flames. The selected initial pressures were between 10 and 40 atm for ignition and 1 atm for the laminar flame while an emphasis was given in all cases to fuel lean conditions ($0.5 < \varphi < 1.0$).

As it was explained in Section 2, the simplification process relies on the multiscale analysis of the full solution of the detailed mechanism. Obviously, the size of the produced skeletal depends on the size of the full solution of the detailed mechanism which can be composed of sets of solutions for different initial conditions. Hence, larger full solution or more sets of solutions will produce larger skeletal mechanisms. An efficient way to tackle this is by selecting representative sets of initial conditions in the targeted parametric space. Although multiple combinations were tested, the most efficient one were found to be the following three sets: $T(0) = 1,200$ K, $p(0) = 25$ atm, $\varphi = 0.7$, $T(0) = 1,500$ K, $p(0) = 25$ atm, $\varphi = 0.7$ and $T(0) = 1,800$ K, $p(0) = 25$ atm, $\varphi = 0.7$, i.e., varying solely the initial temperature. This is not a surprising finding because the chemical pathways are strongly dependent on temperature and only weakly dependent on pressure while stoichiometry does not play considerable role when it is only varied in a small range.

The next step in the process is the determination of the initial kernel of species that will serve as the starting point for the identification of all the important reactions and subsequently species. Multiple combinations were tested and the most efficient one was identified to be the set with the major combustion products, i.e., CO_2 and H_2O. In general, by increasing the initial kernel larger skeletals are produced for the same error.

Last step in the simplification process is the determination of the CSP tolerances (absolute and relative) that determine the number of exhausted modes (Valorani et al. 2018). In a case where the timescales develop sufficiently far from each other and a clear timescale gap exists, these tolerances would not affect significantly the outcome, when varied in a reasonable range, i,e, $10^{-1} < tol_{relative} < 10^{-6}$ and $10^{-5} < tol_{absolute} < 10^{-12}$. Figure 3 displays the percentage error of the ignition delay time for various skeletals produced from the same solution ($T(0) = 1,200$ K and 1,800 K) and initial species kernel but with varying tolerances values. It is shown that each set of tolerances can produce skeletals with different accuracy for each set of conditions but in each case (i.e., $T(0) = 1,200$ K and 1,800 K) the selected tolerances share very similar trends in their accuracy. For the selected set of solutions that formed the basis for the production of the skeletals, the following tolerances values were found to produce a 58-species skeletal of excellent accuracy.

The developed 58-species skeletal mechanism can include He and Ar. The whole 60-species mechanism is attached in the current manuscript as Supplementary Material. The accuracy of the new skeletal has been tested in a wide range of initial conditions for the three-component surrogate, namely ignition delay time and flame speed. In particular, Fig. 4 displays the performance of the skeletal in ignition delay time against the detailed model for $1,000\ K < T(0) < 2,000\ K$, $10\ atm < p(0) < 40\ atm$, $0.5 < \varphi < 1.0$. It is shown that the 58-species skeletal performs quite well in the range of 1,100-2,000 K with an error less than 30%, for all pressures and stoichiometries, but overpredicts ignition at low temperatures of $T \approx 1,000\ K$.

The 56-species skeletal mechanism was also tested against its laminar flame speed. As it is shown in Fig. 5 the skeletal approximates remarkably well the detailed mechanism in a wide range of stoichiometry but it does overpredicts the laminar flame speed at fuel rich mixture. The overprediction at these conditions is reasonable because the detailed mechanism solutions used for the skeletal production were all at the fuel lean stoichiometry of $\varphi = 0.7$. Nevertheless, even in fuel rich conditions, the laminar flame speed error can be considered acceptable.

Finally, in Fig. 6, the timescales the develop in the homogeneous adiabatic autoignition of the three component surrogate are shown for various initial temperatures in the case of the produced skeletal. It is shown that the fastest timescale ranges between $O(10^{-9})$ and $O(10^{-11})$ for the unburnt mixture and it is $O(10^{-12})$ after ignition has occurred. These values are important as they will determine the timestep size in an explicit solver as those used in high fidelity simulations. Therefore, the developed 58-skeletal mechanism is not only efficient in terms of its size but also in terms of the associated fastest timescale, which as was shown is suitable for usage in explicit solvers for multidimensional simulations. Notice also the significant increase of the fastest timescale that is achieved through the simplification process: the skeletal fastest timescale is always more than six orders of magnitude larger compared to the respective timescale of the detailed mechanism at the same initial conditions.

CONCLUSIONS

In the current study, the detailed mechamism of Liu et al. (Liu et al. 2019) was used for the systematic analysis of a three component surrogate jet fuel (66.2% n-dodecane, 15.8% n-proplylbenzene, 18.0% 1,3,5,trimethylcyclohexane), suitable for use as conventional aviation fuel (Jet A1), at conditions relevant to gas turbines. The analysis was performed using mathematical tools from computational singular perturbation (CSP) with an emphasis on the chemical pathways and the species that control the system's ignition in a wide range of initial condtions. It was shown hydrogen and CO-related reactions have a key role and as the initial temperature increases, their importance becomes more pronounced from the very early stage of the ignition process. In addition, the $C_2H_4 \rightarrow C_2H_3 \rightarrow CH_2CHO$ pathway plays a secondary but non-negligible role with an importance increasing with initial temperature, favoring the system's explosive dynamics and, thus, promoting ignition. Another side effect of the increase of the initial temperature is that the thermal runway notably suppresses the chemical runway and temperature becomes the variable with the larger effect of all variables to the system's explosive dynamics. Finally, C_2H_4 was identified by the CSP Po to be a species with a key (secondary) role to the system's explosive dynamics but its role is replaced by C_3H_6 and eventually by O_2, as the initial temperature increases.

In the second part of the current work, a 58-species skeletal mechanism was developed using an algorithmic process based on CSP (Valorani et al. 2006b). The developed skeletal was tested in a wide range of initial conditions, including both ignition delay time and laminar flame speed calculations. In all cases, the skeletal approximated the detailed mechanism with very small error, except for relatively low initial temperatures ($T(0) \approx 1,000\ K$) and fuel rich mixtures ($\varphi > 1.6$) which were in any case, out of scope for the current mechanism. Finally, the produced skeletal is ideal for use in CFD applications not only because of its small size but also because of its sufficiently large associated fast timescale, which was found to be between $O(10^{-9})$ and $O(10^{-11})$.

The current work aims to be the first solid step in the systematic dynamics analysis of aero-propulsion fuels, through the obtained fundamental understanding of the underlying physics, thus, eventually contributing to the development of new combustion modes. Therefore, more studies in this area are required as necessitated by the need for an efficient climate change control.

DATA AVAILABILITY STATEMENT

- All data, models, and code generated or used during the study appear in the submitted article.

SUPPLEMENTAL DATA

The chemical kinetics mechanism is available online in the ASCE Library.

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TABLE 1. The reactions providing significant contribution to the generation of the fast time scale $\tau_{e,f}$, during the autoignition of the surrogate fuel at the three selected conditions under study. The numbering of reactions is the one used in (Liu et al. 2019).

#	Reaction
1037	$aC_3H_5+CH_3(+M) \leftrightarrow C_4H_{81}(+M)$
1230	$C_2H_3+CH_3(+M) \leftrightarrow C_3H_6(+M)$
1231	$aC_3H_5+H(+M) \leftrightarrow C_3H_6(+M)$
1258	$aC_3H_4+H \leftrightarrow aC_3H_5$
1277	$aC_3H_5+HO_2 \leftrightarrow C_3H_5O+OH$
1280	$aC_3H_5+O_2 \leftrightarrow CH_2CHO+CH_2O$
1282	$aC_3H_5+O_2 \leftrightarrow CH_3CO+CH_2O$
1301	$C_2H_2+CH_3 \leftrightarrow pC_3H_4+H$
1317	$C_3H_3+O_2 \leftrightarrow CH_2CO+HCO$
1319	$C_3H_3+HO_2 \leftrightarrow aC_3H_4+O_2$
1320	$C_3H_3+HO_2 \leftrightarrow pC_3H_4+O_2$
1322	$C_3H_3+HCO \leftrightarrow pC_3H_4+CO$
1330	$H+O_2 \leftrightarrow O+OH$
1331	$O+H_2 \leftrightarrow OH+H$
1332	$OH+H_2 \leftrightarrow H+H_2O$
1339	$H+OH+M \leftrightarrow H_2O+M$
1341	$H+O_2(+M) \leftrightarrow HO_2(+M)$
1344	$H_2+O_2 \leftrightarrow H+HO_2$
1348	$HO_2+OH \leftrightarrow H_2O+O_2$
1350	$HO_2+HO_2 \leftrightarrow H_2O_2+O_2$
1352	$H_2O_2(+M) \leftrightarrow OH+OH(+M)$
1359	$CO+OH \leftrightarrow CO_2+H$
1377	$CH_2O+O_2 \leftrightarrow HCO+HO_2$
1381	$HCO+M \leftrightarrow H+CO+M$
1385	$HCO+OH \leftrightarrow CO+H_2O$
1386	$HCO+O_2 \leftrightarrow CO+HO_2$
1390	$CH_3+H(+M) \leftrightarrow CH_4(+M)$
1411	$CH_3+HO_2 \leftrightarrow CH_3O+OH$
1413	$CH_3+O_2 \leftrightarrow CH_2O+OH$
1517	$CH_3+CH_3(+M) \leftrightarrow C_2H_6(+M)$
1526	$C_2H_4+H(+M) \leftrightarrow C_2H_5(+M)$
1538	$C_2H_3+H(+M) \leftrightarrow C_2H_4(+M)$
1540	$C_2H_4+H \leftrightarrow C_2H_3+H_2$
1545	$C_2H_4+OH \leftrightarrow C_2H_3+H_2O$
1553	$C_2H_2+H(+M) \leftrightarrow C_2H_3(+M)$
1559	$C_2H_3+O_2 \leftrightarrow CH_2CHO+O$
1573	$C_2H_2+O \leftrightarrow HCCO+H$
1715	$CH_2+CO(+M) \leftrightarrow CH_2CO(+M)$
1730	$HCCO+OH \leftrightarrow CO+HCOH$
1733	$HCCO+O_2 \leftrightarrow CO_2+CO+H$
1791	$A_1C_3H_7+H \leftrightarrow A_1CH_2CH_2CH_2+H_2$
2012	$PXC_6H_{13}(+M) \leftrightarrow C_2H_4+pC_4H_9(+M)$
2488	$PXC_6H_{13}(+M) \leftrightarrow S_2XC_6H_{13}(+M)$
2532	$PXC_9H_{19}+nC_3H_7 \leftrightarrow nC_{12}H_{26}$
2568	$nC_{12}H_{26}+HO_2 \leftrightarrow SXC_{12}H_{25}+H_2O_2$
2569	$nC_{12}H_{26}+HO_2 \leftrightarrow S_2XC_{12}H_{25}+H_2O_2$
2570	$nC_{12}H_{26}+HO_2 \leftrightarrow S_3XC_{12}H_{25}+H_2O_2$
2571	$nC_{12}H_{26}+HO_2 \leftrightarrow S_4XC_{12}H_{25}+H_2O_2$
2572	$nC_{12}H_{26}+HO_2 \leftrightarrow S_5XC_{12}H_{25}+H_2O_2$
2585	$pC_{12}H_{25}O_2 \leftrightarrow p_{12}OOHX_2$
2599	$T_{135}MCH+OH \leftrightarrow CH_3SXD_{35}MCH+H_2O$
2601	$T_{135}MCH+O_2 \leftrightarrow TXT_{135}MCH+HO_2$

TABLE 2. The largest Time scale Participation Indices (TPI) and CSP Pointers (Po) during the autoignition process of the surrogate fuel for the three sets of initial conditions under study; $T(0) = 1,200$ K, $1,500$ K and $1,800$ K. In all cases, $p(0) = 25$ atm, $\varphi = 0.7$. The symbols "f" and "b" stand for forward and backward, respectively. The numbers in the first row correspond to the numbered points in Fig. 2. Reactions in blue/red are hydrogen/CO-related, respectively.

Case	Metric	P1		P2		P3		P4		P5	
		Rxn	TPI/Po	Rxn	TPI/Po	Rxn	TPI/Po	Rxn	TPI/Po	Rxn	TPI/Po
1,200 K $t_{ign} = 3.4987E-04$ s	t $[s]$	0		8.76×10^{-4}		1.75×10^{-4}		2.62×10^{-4}		3.49×10^{-4}	
	τ_e $[s]$	2.29×10^{-5}		4.47×10^{-4}		1.85×10^{-4}		7.60×10^{-5}		1.15×10^{-6}	
	TPI	1352f	8.33 %	1330f	3.81 %	1545f	5.24 %	1330f	6.51 %	1330f	23.44 %
		1330f	3.63 %	1545f	3.73 %	1330f	4.81 %	1545f	5.97 %	1381f	4.10 %
		2570f	2.40 %	1341f	3.14 %	1277f	3.64 %	1277f	3.71 %	1341f	-4.07 %
		2572f	2.40 %	1277f	2.78 %	1350f	-3.03 %	1350f	-2.91 %	1348f	-3.57 %
		2568f	2.36 %	1350f	-2.65 %	1341f	2.69 %	1348f	-2.48 %	1332f	3.51 %
		1413f	2.35 %	1517f	-2.37 %	1526b	2.18 %	1559f	2.65 %	1730f	-3.29 %
		2585f	2.34 %	1413f	2.34 %	1559f	2.10 %	1341f	2.60 %	1390f	-2.61 %
		2569f	2.30 %	1037f	-2.31 %	1037f	-2.08 %	1526b	2.30 %	1573f	2.23 %
		2571f	2.20 %	1411f	-1.66 %	1413f	1.90 %	1352f	2.05 %	1344b	-2.17 %
		2599f	-1.89 %	1526b	1.53 %	1517f	-1.88 %	1037f	-1.94 %	1545f	2.17 %
	Po	HO_2	0.47	T	0.54	T	0.65	T	0.81	T	0.90
		H_2O_2	0.24	C_2H_4	0.27	C_2H_4	0.12	C_2H_4	0.05	O_2	-0.06
		CH_3	0.12	HO_2	0.24	HO_2	0.81	HO_2	0.03	OH	-0.05
1,500 K $t_{ign} = 1.46248E-05$ s	t $[s]$	0		3.66×10^{-6}		7.31×10^{-6}		1.10×10^{-5}		1.46×10^{-5}	
	τ_e $[s]$	3.85×10^{-5}		8.56×10^{-6}		6.52×10^{-6}		3.31×10^{-6}		2.92×10^{-7}	
	TPI	1330f	4.82 %	1330f	11.94 %	1330f	14.73 %	1330f	18.41 %	1330f	24.49 %
		1280f	3.91 %	1545f	6.20 %	1545f	5.76 %	1545f	5.10 %	1359f	10.89 %
		1413f	3.88 %	1231f	-3.27 %	1231f	-3.24 %	1348f	-3.21 %	1330b	-6.90 %
		1320b	3.82 %	1341f	3.20 %	1559f	2.82 %	1559f	3.16 %	1332f	5.94 %
		1341f	2.73 %	1277f	3.15 %	1348f	-2.59 %	1231f	-2.00 %	1331f	4.74 %
		1231f	-2.69 %	1559f	2.28 %	1540f	1.79 %	1540f	1.75 %	1341f	-4.42 %
		1282f	2.55 %	1350f	-2.27 %	1277f	1.74 %	1553b	-1.60 %	1322b	-3.90 %
		1377f	2.53 %	1381f	-1.97 %	1341f	1.72 %	1344b	-1.59 %	1385f	-3.20 %
		1386f	2.17 %	1386f	1.82 %	1350f	-1.59 %	1573f	1.57 %	1339f	-2.91 %
		1381f	-2.16 %	1348f	-1.82 %	1381f	-1.46 %	1301b	-1.34 %	1331b	-2.41 %
	Po	$NC_{12}H_{26}$	-1.11	T	0.66	T	0.82	T	0.89	T	0.83
		T	-0.57	CH_2O	0.06	C_3H_6	0.09	C_3H_6	0.08	H_2	-0.43
		$T_{135}MCH$	0.56	C_2H_4	0.06	O_2	-0.06	O_2	-0.06	O	0.40
1,800 K $t_{ign} = 1.4658E-06$ s	t $[s]$	0		3.65×10^{-7}		7.35×10^{-7}		1.10×10^{-6}		1.47×10^{-6}	
	τ_e $[s]$	9.87×10^{-6}		9.47×10^{-7}		6.29×10^{-7}		3.19×10^{-7}		1.71×10^{-7}	
	TPI	1330f	12.95 %	1330f	20.63 %	1330f	20.63 %	1330f	22.14 %	1330f	24.84 %
		1386f	3.35 %	1545f	3.52 %	1545f	2.86 %	1381f	4.42 %	1359f	8.21 %
		1381f	-3.28 %	1559f	2.97 %	1559f	2.53 %	1730f	-4.19 %	1332f	7.26 %
		1540f	3.03 %	1553b	-2.63 %	1573f	2.41 %	1332f	3.11 %	1331f	5.74 %
		1553b	-2.68 %	1231f	-2.62 %	1381f	2.34 %	1573f	2.74 %	1330b	-5.71 %
		1573f	2.46 %	1348f	-2.07 %	1730f	-2.30 %	1538f	-2.35 %	1322b	-4.38 %
		1413f	2.19 %	1573f	1.87 %	1348f	-1.99 %	1733f	2.00 %	1385f	-3.63 %
		1341f	2.06 %	1540f	1.77 %	1733f	1.73 %	1344b	-1.84 %	1730f	-3.12 %
		1559f	1.78 %	1344b	-1.46 %	1344b	-1.66 %	1545f	1.80 %	1331b	-2.92 %
		1532f	1.70 %	1730f	-1.45 %	1332f	1.57 %	1348f	-1.62 %	1341f	-2.89 %
	Po	$T_{135}MCH$	0.98	T	0.78	T	0.76	T	0.73	T	0.73
		$NC_{12}H_{26}$	-0.83	C_3H_6	0.09	O_2	-0.07	O_2	-0.08	H_2	-0.43
		C_2H_4	0.81	O_2	-0.08	C_3H_6	0.06	H	0.07	O	0.42

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(a)(b)(c)

Fig. 1. The developing timescales during the homogeneous adiabatic constant pressure autoignition of the 3-component surrogate at $T(0) = 1,200$ K (a), $T(0) = 1,500$ K (b) and $T(0) = 1,800$ K (c). Solid and dashed lines represent dissipative and explosive timescales, respectively. The dotted line denotes the temperature evolution while the circle signs denote the $M + 1$-timescale. In all cases, $\varphi = 0.7$ and $p(0) = 25$ atm.

(a)(b)(c)

Fig. 2. The evolution of the explosive timescales (solid and dashed lines) and temperature (dotted lines) during the homogeneous adiabatic constant pressure autoignition of the surrogate fuel at $T(0) = 1,200$ K (a), $T(0) = 1,500$ K (b) and $T(0) = 1,800$ K (c). P_1-P_5 represent the points that CSP diagnostics were generated. Solid and dashed lines represent the fast ($\tau_{e,f}$) and the slow ($\tau_{e,s}$) explosive timescales, respectively. In all cases, $\varphi = 0.7$ and $p(0) = 25$ atm.

(a) (b)

Fig. 3. Comparison of the ignition delay error against total number of species for various tolerances values at: (a) $T(0) = 1,200$ K and (b) $T(0) = 1,800$ K. In all cases, $p(0) = 25$ atm, $\varphi = 0.7$.

(a)(b)(c)

Fig. 4. Comparison of the ignition delay times between the detailed (solid lines) and the 58-species skeletal (dashed lines) kinetics mechanisms at various initial conditions (homogeneous adiabatic constant pressure autoignition); (a) $\varphi = 0.5$, (b) $\varphi = 0.7$, (c) $\varphi = 1.0$.

(a)(b)

Fig. 5. Comparison of the laminar flame speed between the detailed (solid line) and the 58-species skeletal (dashed line) kinetics mechanisms for the 3-component surrogate at various stoichiometries; (a) $T_u = 300\ K$, (b) $T_u = 470\ K$. In all cases, $p = 1$ atm.

(a)(b)(c)

Fig. 6. The developing timescales of the 58-species skeletal mechanism during the homogeneous adiabatic constant pressure autoignition of the 3-component surrogate at $T(0) = 1,200$ K (a), $T(0) = 1,500$ K (b) and $T(0) = 1,800$ K (c). The dotted line denotes the temperature evolution. In all cases, $\varphi = 0.7$ and $p(0) = 25$ atm.