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Low Temperature Autoignition of Conventional Jet Fuels and Surrogate Jet Fuels with Targeted Properties in a Rapid Compression Machine

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Abstract

The autoignition characteristics of conventional jet fuels (category A) and alternative fuels with targeted properties (category C) are investigated using a rapid compression machine and the direct test chamber charge preparation approach. The category C fuels were purposefully built to anticipate special property variations that generally occur in alternative fuels. Ignition delay measurements were made to examine the effects of these unique fuels at low compressed temperatures ($625 \text{ K} \leq T_c \leq 735 \text{ K}$), a compressed pressure of $P_c = 20 \text{ bar}$ and equivalence ratios of $\phi = 0.25, 0.5$ and 1.0 in synthetic dry air. Chemical make-up of the fuel shows insight into the effect of the amount of branching in isoalkanes and aromatic influences on autoignition. The results show noteworthy variability in the ignition properties at these low temperature and lean conditions. This variability may impact combustion performance when the engine is running outside the normal operational map or for new engine architectures in the future.

Keywords: Autoignition; Rapid Compression Machine; Category A Fuel; Category C Fuel; Low Temperature Combustion

1. Introduction

Increasing difficulties in the extraction of crude oil and concerns over energy security have resulted in a growing interest in the use of alternative fuels. Both military and commercial sectors desire that any alternative fuel be a drop-in replacement, requiring no retrofitting of current combustion devices to accommodate new fuels. Recently, a large number of alternative jet fuels from various sources have been widely introduced into the market. The Federal Aviation Administration Center of

Excellence (ASCENT), in collaboration with the Air Force Research Laboratory, has established the National Jet Fuel Combustion Program (NJFCP), which has tasked itself with determining the suitability of alternative fuel options. Within the program, the tested fuels have been divided into three general categories: A, B, and C. Category A fuels are the current petroleum-based fuels used in commercial and military applications. Category B fuels are alternative fuels that have already undergone strenuous analysis and testing. Category C fuels are alternatives with little to no testing, but also include purposefully built surrogates made to anticipate special property variations that generally occur in alternative fuels, such as narrow boiling range, limited components, and blending effects, whose impacts are poorly understood [1]. Chemical kinetic data for these fuels in the form of ignition delay measurements is a key piece of information needed for verification and aid in developing accurate simulations. However, a lack of data has created a push for the characterization of the ignition properties of new alternative fuels to support integration of these fuels into existing systems. Recent studies include those of Fischer-Tropsch (FT), hydroprocessed renewable jet (HRJ), alcohol to jet (ATJ), and direct sugar to hydrocarbon (DSHC) fuels [2-10] and references therein.

The objective of this work is to provide data investigating the autoignition characteristics of category C jet fuels and to compare their autoignition properties against conventional aviation (category A) fuels under low-temperature (<750K) conditions in a rapid compression machine (RCM). Ignition delay measurements provided in this study will be important for validation of kinetic models, while examination of ignition delay chemistry based on chemical structures will enhance understanding of combustion; both of which will contribute toward the successful integration of next-generation bio-derived fuels with existing aviation systems.

2. Experimental Methods

2.1 Test Fuels and Solvents

The focus of this study is to compare the combustion properties of category C fuels to the currently used category A fuels. Three category A fuels: land-based jet propellant, JP-8 (A-1), a commercial airline and soon-to-be military fuel, Jet A (A-2), and sea-based jet propellant, JP-5 (A-3) are included in this study. In addition, the study will examine six category C fuels: a highly branched isoparaffinic ATJ which consists primarily of isododecane and isocetane (C-1), a “bimodal” blend of 84% vol. tetradecane and 16% vol. 1,3,5-trimethylbenzene (C-2), blend of 64% vol. A-3 and 36% vol. farnesane (C-3), blend of 40% vol. C-1 and 60% vol. high-temperature FT isoparaffinic kerosene (C-4), blend of 73% vol. decane and 27% vol. 1,3,5-trimethylbenzene (C-5), and a DSHC fuel that contains 2,6,10-trimethyl dodecane (farnesane). For reference, a summary of the fuel blends studied is provided in Table 1. All fuels were obtained from the U.S. Air Force Research Laboratory. As previously

mentioned, category C fuels have unique features that will be described briefly, although they are not fully understood. C-1 represents a low cetane fuel with an unusual boiling range, while also exhibiting highly branched species for analysis. C-2 will highlight the issue of potential dominance of particular fuel chemistry in the more volatile part of the boiling range, in this case, C9 aromatics [1]. C-3 allows for the examination of a high viscosity fuel (8.3 cSt at -20°C). C-4 represents a wide-boiling blend with a low cetane number because of the presence of C-1, which contains highly branched alkanes [11]. C-5 will examine a narrow boiling fuel, in the lower boiling range of conventional jet fuels. Farnesane is proposed as a blending component for use with category A and C fuels. These unique features of the category C fuels are more clearly displayed in Fig. 1 and Table 2, which are explained further below.

Table 1. Category C Fuels Composition

Fuel	Blend Composition (% vol.)
C-1	100% highly branched ATJ Fuel
C-2	84% tetradecane & 16% 1,3,5-TMB
C-3	64% Jet A-3 & 36% farnesane
C-4	60% isoparaffinic FT & 40% Jet C-1
C-5	73% decane & 27% 1,3,5-TMB

Figure 1 shows normalized total ion chromatograms of the category A and C fuels being investigated. Comparison of the category A fuels shows that these petroleum fuels all have a very similar structure with prominent n-alkane peaks. One major difference that can be seen between the category C fuels in comparison to the category A fuels is the overall lack of normal alkanes. A lack of normal alkanes may be an indication that the ignition delay of the fuels will be longer, unless significant amounts of lightly branched alkanes are present. C-1 and farnesane both are comprised of very few species, thus have very small boiling ranges and present a stark contrast from the category A fuels. C-5 also has a very small boiling range that is on the lower boiling range of conventional jet fuel. The remainder of the category C fuels have larger boiling ranges, but contain the dominant peaks of the neat components in their blend composition, while the category A fuels contain a more Gaussian shaped carbon number distribution.

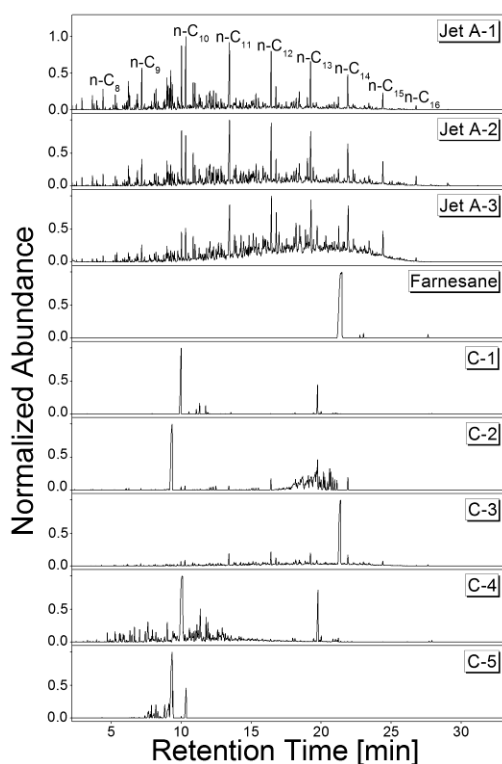


Figure 1. Total ion chromatograms of all test fuels.

Combustion issues with alternative fuels mainly exist because of the intrinsic nature of the fuels. Table 2 displays selected chemical and physical properties of the category C fuels in comparison to the conventional fuels. One challenge associated with using synthetic jet fuels as drop-in replacements for conventional jet fuels is achieving the fuel density specifications set for military fuels (0.775 – 0.840 g/mL). Table 2 shows that only the C-2 and C-3 fuels meet the density specification, while the remainder of the category C fuels are just below the acceptable minimum of the required fuel density specification. Another feature to note is that category C fuels in general contain a much higher ratio of isoalkanes to normal alkanes, which was a feature also noted in Fig. 1. The aromatic content of the category C fuels is considerably varied compared to that of the category A fuels. C-1, C-4, and farnesane contain almost no aromatics, C-5 has a markedly higher aromatic content than the category A fuels, and C-2 and C-3 fall within the aromatic content range of the category A fuels. Aromatics are vital because of their role in the proper swelling of elastomer seals and are commonly linked to volumetric energy density, which is important when considering power and/or range of the combustion device [12, 13]. The disparity between the fuels and the uniqueness of their properties, as marked in their cetane numbers, motivates investigation into the autoignition characteristics of the category C fuels under different test conditions.

Table 2. Chemical and Physical Properties of Category A and C Fuels

Fuel	POSF	Cetane	Flash Pt. (°C)	Density (g/mL)	n-alkane (%wt)	isoalkane (%wt)	cycloalkane (%wt)	aromatic (%wt)
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A-1	10264	40.4	42	0.780	26.82	39.69	20.08	13.41
A-2	10325	47	48	0.803	20.03	29.45	31.86	18.66
A-3	10289	47.9	60	0.827	13.89	18.14	47.39	20.59
Farnesane	10320	-	110	0.770	0	100	0	0
C-1	10262	17.1	50	0.760	<0.01	99.62	0.05	<0.01
C-2	12223	50.4	40	0.782	5.16	77.51	0.07	17.05
C-3	12341	47.0	63	0.808	9.17	45.19	31.72	13.61
C-4	12344	28.0	108	0.760	0.23	98.94	0.43	0.39
C-5	12345	39.6	118	0.770	17.66	51.58	0.07	30.68

2.2 Heated Rapid Compression Machine

The main source of data collection for this study involved the use of a RCM, which is depicted in Fig. 2. A brief description of the RCM will follow, while a brief summary of its capabilities can be seen in Table 3, for full details refer to Allen et al. [14, 15]. The RCM is a pneumatically driven, hydraulically stopped apparatus that simulates a single compression stroke of a diesel engine. The fuel charge preparation method utilized is known as the direct test chamber (DTC) approach and is used as described by Allen et al. [15]. Tersely, DTC works by first preloading the desired amount of oxidizer into the combustion cylinder. A fuel injector mounted directly into the RCM then injects a precise amount of fuel to achieve a specified equivalence ratio. Sufficient time is then allowed for the fuel to reach and maintain spatial homogeneity via a controlled heating system prior to compressing the gaseous mixture.

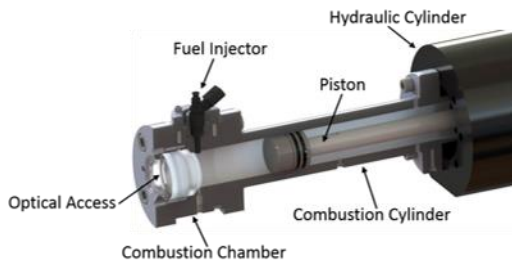


Figure 2. Cut-away isometric side view of the RCM at the University of Illinois at Urbana-Champaign.

Table 3. Summary of RCM Capabilities and Current Study Parameters

Driving, Stopping Mechanism	Compression Ratio	Key Features	Pc (bar)	Tc (K)
Pneumatically driven, hydraulically stopped	4 – 17 (creviced)	Creved piston, optical access, adjustable stroke and clearance volume	20	625 - 735

2.3 RCM Data Analysis and Experimental Uncertainty

The ignition delay is defined as the amount of time required for a fuel to autoignite under compressed conditions. In order to accurately determine the ignition delay, a standard definition is required so all tests are compared on an equivalent basis.

The starting time of the ignition delay is determined at top dead center (TDC) of the piston stroke, while the endpoint is when peak pressure release occurs. The starting point is easily found from the pressure profiles recorded during tests in the RCM, while the start of combustion can be found using the first derivative of the pressure trace, where maxima values correspond to ignition points. The overall ignition delay (τ) is defined as $\tau = \tau_1 + \tau_2$, where τ_1 represents the first stage delay and τ_2 is the time elapsed from the first stage ignition until the main combustion event occurs. This definition allows for multistage ignition to be accounted for which is common in low temperature combustion and at reduced equivalence ratios.

Compressed gas temperatures, T_c , are calculated at TDC and used as the reference temperature for comparing ignition delay experiments. A creviced piston, based on the design of Mittal et al. [16] was used to eliminate significant boundary roll-up issues. LIF experiments conducted by Mittal et al. using the creviced piston showed stabilized temperature conditions axially across the core in the RCM for up to 114 ms after compression under elevated pressures, thereby mitigating heat loss and cooling effects during and after compression. Thus, an adiabatic core post-compression is achieved, allowing the calculation of T_c by the relationship, $\int_{T_0}^{T_c} \frac{\gamma}{\gamma-1} \frac{dT}{T} = \ln \frac{p_c}{p_0}$, where T is temperature, P is pressure, γ is the temperature-dependent specific heat ratio, and the subscripts 0 and c correspond to initial and compressed conditions, respectively. One condition to solve for T_c , is that the temperature-dependent specific heat ratios for all gas phase species must be known; however, the exact compositions of the fuels tested is undetermined. In absence of this data, the thermodynamic properties were approximated for all the fuels, with the exception of C-14, using surrogate approximations formulated by Xu et al. [17]. The thermodynamic data was provided from pure component surrogates created with specific attention paid to matching the molecular weights of the category A and C fuel of interest. For farnesane, T_c was calculated using the thermophysical properties of n-dodecane with a correction factor based on farnesane's molecular weight ratio to n-dodecane. The thermophysical property data of n-dodecane was found via Burcat [18].

A minimum of three ignition delay measurements have been tested at each temperature condition and the standard deviations of the measurements are represented by error bars when presenting results in this paper. The main uncertainty taken into account results from calculating compressed temperature, which depends on uncertainties in initial temperature (± 3 K), initial pressure ($\pm 0.08\%$), initial volume (± 0.004 L), compressed pressure (± 0.3 bar), injected fuel mass ($\pm 3\%$), and thermophysical property data ($\pm 0.5\%$). These inputs have been used to calculate the uncertainty in the compressed temperature as $\pm 1.25\%$ (8 – 9 K), which is represented by error bars in subsequently presented data.

3. Results and Discussion

Ignition delay measurements of the category A (A-1, A-2, A3) and category C (C-1, C-2, C-3, C-4, C-5, farnesane) fuels were studied at low compressed temperatures of $625 \text{ K} \leq T_c \leq 735 \text{ K}$, a compressed pressure of 20 bar and equivalence ratios of $\phi = 0.25, 0.5$ and 1.0 in synthetic dry air (79% N_2 , 21% O_2).

3.1 Comparison of Category A Fuels

The main goal of this study is to investigate the autoignition behavior of the category C fuels to observe the effects of their unique characteristics on combustion. However, in order to make a comparison, the category A fuels must first be examined. Figure 3 displays the ignition delays of the category A fuels at a $P_c = 20$ bar and $\phi = 1.0, 0.5$, and 0.25 . The fuels all show a negative dependence on equivalence ratio and for a given test condition, the ignition delay is longer at lower equivalence ratios. Onset of negative-temperature-coefficient (NTC) behavior is apparent at the higher temperature conditions of the $\phi = 0.5$ case, which is marked by a decrease in reaction activity despite an increase in temperature. The overall ignition delays are very similar among category A fuels, with the largest difference apparent at the leanest condition, where Jet A-3 has the longest delay. However, for all conditions examined, the ignition delays are considered nearly identical based on experimental error. Conventional kinetic modeling of alkanes suggests that reactivity is controlled by low-temperature chain branching: $\text{R} + \text{O}_2 \leftrightarrow \text{RO}_2 \leftrightarrow \text{QOOH} (+\text{O}_2) \leftrightarrow \text{OOQOOH} \rightarrow 2\text{OH} + \text{products}$ [19]. The rate of this reaction pathway is mostly governed by the $\text{R} + \text{O}_2 \leftrightarrow \text{RO}_2$ equilibrium reaction and the rate of isomerization $\text{RO}_2 \leftrightarrow \text{QOOH}$. Therefore, the similarity of category A fuel compositions based on the GC results yields an expected likeness in reactivity in the low temperature chemistry pathway. Because of the similarity in category A fuels, comparisons of combustion for category C fuels will be made to the best-case category A fuel, Jet A-1; however, fuel blends that contain any of the other category A fuels will use that fuel for comparison instead.

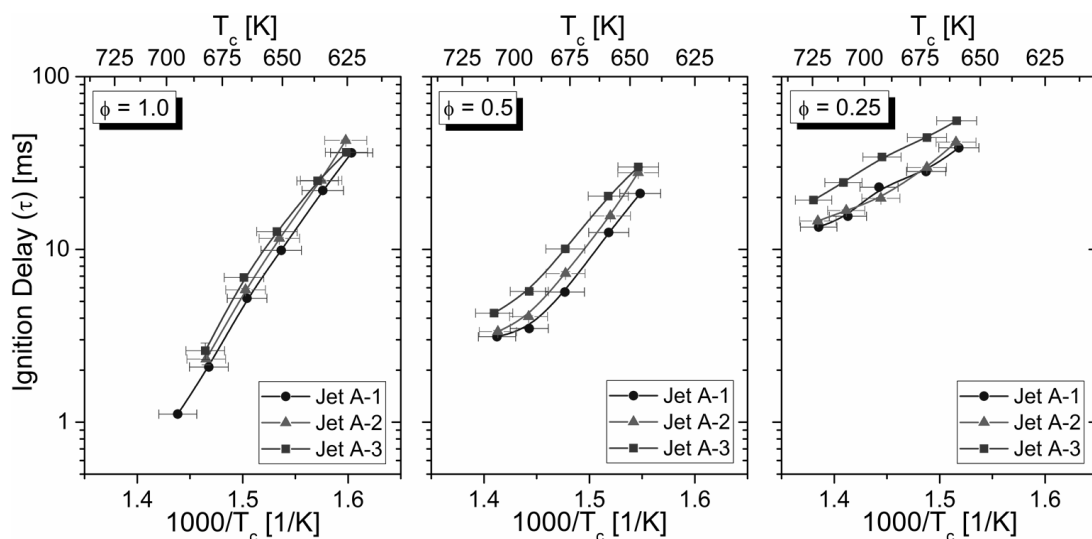


Figure 3. Ignition delays for category A fuels at $P_c = 20$ bar and $\phi = 1.0, 0.5,$ and 0.25 .

3.2 Comparison of Jet A-1, Jet C-1, and Jet C-4

Figure 4 displays low-temperature RCM data for Jet A-1, C-1, and C-4. Inspecting the neat C-1 and conventional fuel ignition delays shows C-1 has a longer ignition delay, that drastically increases at the $\phi = 0.5$ condition. Data for C-1 at the $\phi = 0.25$ condition could not be obtained since no discernible ignition occurred within the conducted test runtime. The long ignition delays of C-1 are attributed to the significantly branched nature of isododecane and isocetane, which has been shown to result in retarded ignition time [20, 21]. The reasoning for this is that the high levels of branching can create issues with isomerization reactions, due to a lack of secondary C-H bonds, thereby inhibiting the energetically favorable six-membered ring transitions under the low-temperature chemistry pathway. Additionally, isocetane's chemical structure, which is similar to that of isooctane, has been shown to react and produce a high level of isobutene as an intermediate that is unusually stable, also attributing to the longer ignition delay [21].

Investigation of Jet C-4 in Fig. 4 shows similar ignition delays at $\phi = 1.0$ relative to the conventional fuels, with differences becoming more apparent at increased temperatures, where C-4 appears to have an earlier entrance into the NTC region noted by the decrease in reactivity. Since C-4 is a blend of C-1 and a high temperature FT isoparaffinic kerosene, examining the chemistry of each fuel individually could provide insight into the combustion behavior observed. Valco et al. [3] previously examined the FT fuel and found very similar ignition delays compared to conventional jet fuels despite the lack of aromatic content in the fuel, with early entrance to NTC being noted at reduced equivalence ratios. Since the FT fuel is almost completely comprised of isoparaffins, the branching is hypothesized to be moderate where enough secondary hydrogens are available to provide $RO_2 \leftrightarrow QOOH$ isomerization rates similar to those found in the category A fuels. Therefore, the longer ignition delays,

particularly at reduced equivalence ratios, are attributed to the addition of the previously discussed C-1, whose heavy branching and isobutene formation causes the stark difference between Jet C-4 and A-1, particularly at reduced equivalence ratios.

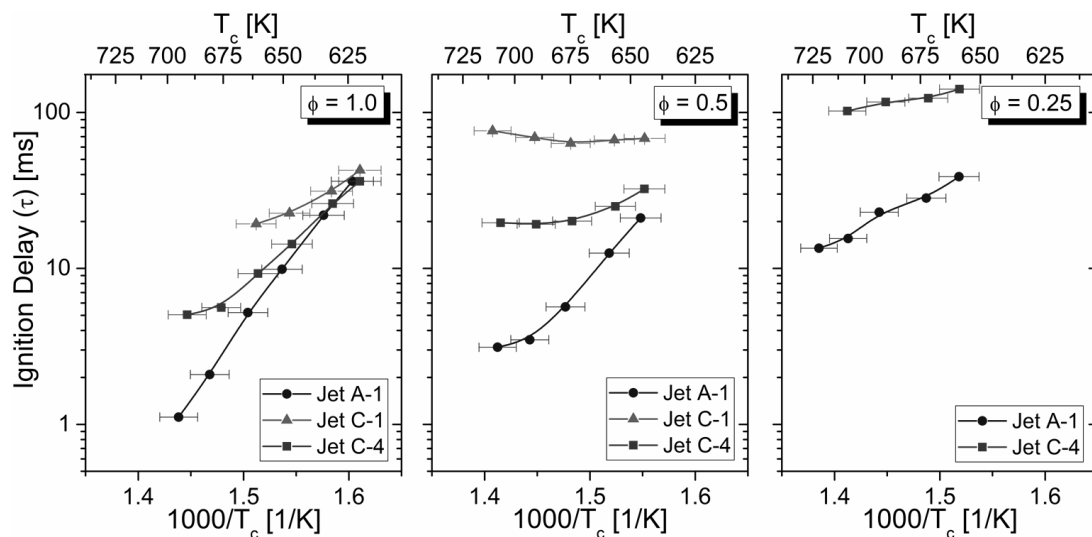


Figure 4. Ignition delays for Jet A-1, Jet C-1, and Jet C-4 at $P_c = 20$ bar and $\phi = 1.0, 0.5,$ and 0.25 .

3.3 Comparison of Jet A-1, Jet C-2, and Jet C-5

Ignition delay measurements for Jet C-2 and C-5, as well as Jet A-1 for comparison, are presented in Fig. 5. Jet C-2 has very similar ignition delays to conventional fuel under all conditions studied, only showing slightly faster ignition versus the conventional fuel in the leanest condition. The composition of C-2 is largely C14 range isoalkanes with small concentrations of n-alkanes and cycloalkanes, but also contains aromatic content similar to that found in conventional fuels. As a result, the composition and average molecular weight is akin to that of conventional fuels, which likely results in the similar ignition delays found, since the fuels are expected to undergo a similar kinetic pathway. Slight differences in the ignition delays are more apparent at $\phi = 0.25$ where a somewhat shorter delay is found for C-2, which is likely the result of differences in the NTC behavior of the fuels, which introduces additional branching pathways.

Examination of Fig. 5 illustrates that C-5 has a longer ignition delay under all conditions studied in comparison to Jet A-1. C-5 investigates the effects of a narrow boiling range fuel which comprises C9 aromatics at higher content than conventional jet fuels and C10 isoalkanes, with small amount of n-alkanes and cycloalkanes. A longer ignition delay for C-5 may be the result of two factors. The first reason is shorter carbon chains, which have slower ignition delays than longer chained hydrocarbons, though studies have shown this has limited effect [22]. Therefore, the more likely cause is higher aromatic content (30% vol), which has been connected to a loss in reactivity under low temperature conditions due to the buildup of

large amounts of stabilized benzylic radicals because of the favorable initial hydrogen abstraction pathway that scavenges available radicals, thereby slowing combustion chemistry [23].

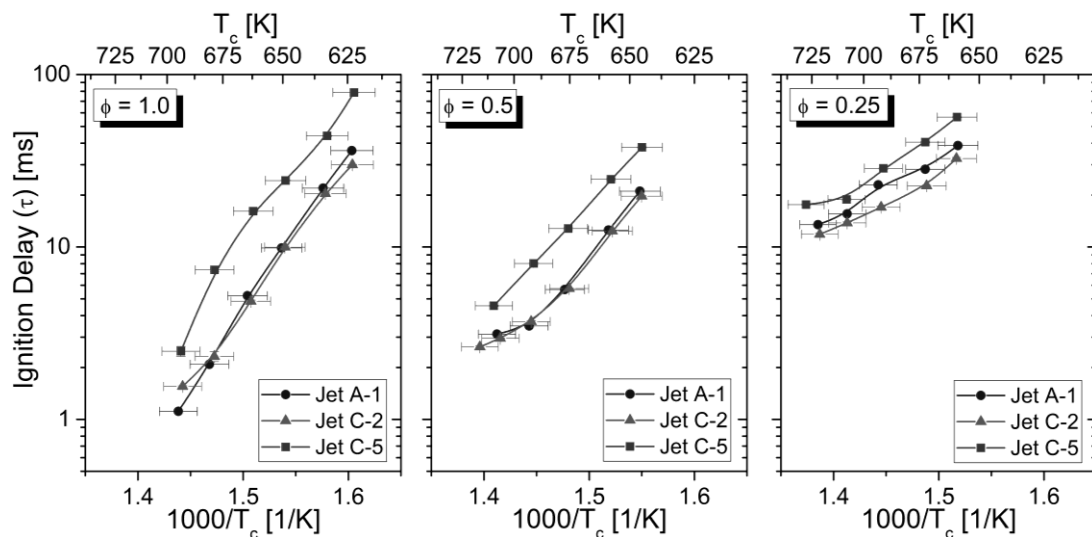


Figure 5. Ignition delays for Jet A-1, Jet C-2, and Jet C-5 at $P_c = 20$ bar and $\phi = 1.0, 0.5,$ and 0.25 .

3.4 Comparison of Jet A-3, Jet C-3, and Farnesane

Figure 6 displays the low-temperature ignition delay data for Jet A-3, C-3, and farnesane. Examination of farnesane shows shorter ignition delays than both Jet A-3 and C-3. Farnesane is a long chain C15 hydrocarbon with minimal branching and as a result, reacts rapidly in low-temperature conditions because of ample secondary hydrogens available, allowing for energetically favorable six-membered ring transitions to aid in isomerization pathways. Farnesane exhibits reduced ignition delays across all equivalence ratios, maintaining a significant reduction in ignition delay even as the ignition delays for Jet A-3 and C-3 converge in the leanest case at $\phi = 0.25$. Thus, the combined effects of high carbon number (long chain HC) and light branching can be considered significant for low temperature conditions for lean to stoichiometric mixtures.

The results for C-3, a blend of Jet A-3 and farnesane, show that it falls between the ignition delays of the two neat fuels, which is expected since the category A fuels all have a similar delay. Thus, adding a significant portion of farnesane, a long chain hydrocarbon with minimal branching, should aid in shorter ignition delays. The leanest condition of $\phi = 0.25$ shows an interesting convergence in the ignition delay of the blend, C-3, to Jet A-3. This behavior has been noted in the past by Valco et al. [3], where the neat fuels tend to maintain the same difference in reactivity at all equivalence ratios; however, the blends do not maintain this reactivity difference and converge toward a parent fuel.

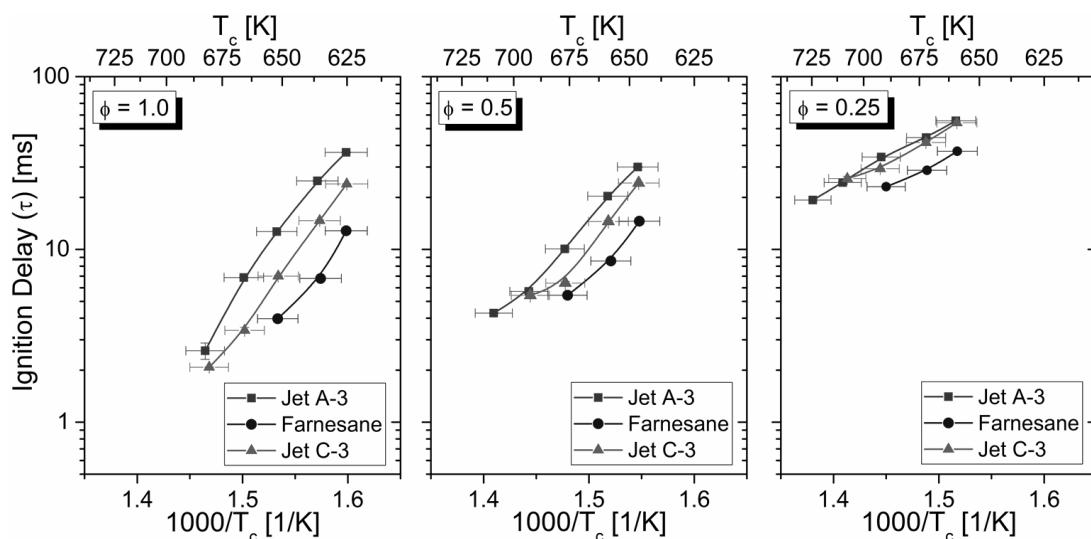


Figure 6. Ignition delays for Jet A-3, Jet C-3, and farnesane at $P_c = 20$ bar and $\phi = 1.0, 0.5,$ and 0.25 .

4. Conclusions

In this study, the autoignition characteristics of three conventional fuels (A-1, A-2, and A-3) and six category C fuels (C-1, C-2, C-3, C-4, C-5, farnesane) have been investigated utilizing a RCM with the DTC method under low-compressed temperatures (below 750K), at a compressed pressure of 20 bar, and at stoichiometric and fuel-lean conditions of $\phi = 1, 0.5,$ and 0.25 . Examination of the chemical composition of each fuel and the corresponding effect on the ignition behavior was inferred from ignition delay results. Specific notable behaviors include the effect of branching in isoparaffins where low levels of branching showed limited effects, while significant branching showed a strong reduction in reactivity. In addition, the influence of aromatic structure on ignition delay was apparent with increased delays due to radical scavenging. The results of this study will provide valuable data for current and future kinetic models that will aid in current and future engine designs. In addition, this study shows the need to further investigate chemical structure and multi-component fuel composition on ignition chemistry, especially under low temperature and lean conditions of disparate fuels, to enhance combustion chemistry understanding and improvement of kinetic models.

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Figure Captions

Figure 1. Total ion chromatograms of all test fuels.

Figure 2. Cut-away isomeric side view of the RCM at the University of Illinois at Urbana-Champaign.

Figure 3. Ignition delays for category A fuels at $P_c = 20$ bar and $\phi = 1.0, 0.5,$ and 0.25 .

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Figure 5. Ignition delays for Jet A-1, Jet C-2, and Jet C-5 at $P_c = 20$ bar and $\phi = 1.0, 0.5,$ and 0.25 .

Figure 6. Ignition delays for Jet A-3, Jet C-3, and farnesane at $P_c = 20$ bar and $\phi = 1.0, 0.5,$ and 0.25 .