Exhaust gas recirculation (EGR) and other dilution strategies are powerful tools under consideration today to reduce air toxic emissions and improve combustion performance. Understanding the kinetic and thermal effects of EGR in an engine can be difficult, particularly when chemistry and fluid physics interactions are significant. Alternatively, experiments that attempt to isolate combustion chemistry, such as ignition studies, often vary the buffer gas to control the experimental conditions. This work explores the kinetic and thermal effects of diluent gas composition on autoignition at concentrations relevant to internal combustion engines, rapid compression machines, and shock tubes. Pressure-time histories were computed with CHEMKIN software for a closed 0-D homogeneous reactor under adiabatic and constant volume conditions for the two primary reference fuels, iso-octane and n-heptane, and for a representative alcohol, n-butanol. Stoichiometric mixtures, $\phi = 1.0$, and a temperature range of 600-1100 K were considered. The alkanes were studied at initial pressures of 9.0 atm and 60.0 atm, and the alcohol was studied at initial pressures of 3.2 atm and 60.0 atm. The fuels and simulation conditions were selected based on the relevance to engine operating conditions and previously published experimental studies, including data from the University of Michigan rapid compression facility. Dilution of 5.64:1 (mole basis) and 3.76:1 of buffer gas to $O_2$ were considered. Diluent gases considered in this study were argon, nitrogen, water, and carbon dioxide. The model results were characterized by ignition delay times, pressure-time histories, and heat release rates. The heat release rates were sensitive to diluent composition with carbon dioxide exhibiting relatively high levels of early and late heat release relative to the other buffer gases. Local sensitivity analysis of the third-body collision efficiencies for the diluent gases indicated that low-temperature combustion was particularly sensitive to the effects of buffer gas composition. This study highlights the significance of buffer gas composition on the uncertainties in low-temperature combustion chemistry, particularly $H_2O_2$ and $HO_2$ decomposition and recombination reactions. Buffer gas composition was found to impact the ignition delay time and heat release rates most in the negative temperature coefficient (NTC) region for the fuels studied (with changes of greater than a factor of 2 in ignition delay time and heat release rate). Outside the NTC region, the effects of changes in buffer gas composition were small (<20%).

1. Introduction

Dilution strategies, such as exhaust gas recirculation (EGR), are important tools to achieve high efficiency, low-emission combustion. There are multiple mechanisms by which EGR can reduce exhaust NOx and particulate matter, including thermo-physical cooling, compositional dilution, and direct kinetic interactions through three-body reactions. These kinetic and thermal effects are also important in that they can play significant roles on reaction rates, autoignition times, and heat release rates. Internal combustion engines are not well-suited for separating these thermal and kinetic effects as they can suffer from competing kinetic and fluidic time scales. Despite these challenges, there have been valuable experimental and computational ICE studies that have investigated the thermal and kinetic effects of buffer gases on autoignition or combustion phasing and product formation [1, 2, 3, 4, 5].

On a related note, buffer gas composition is used to control state conditions in combustion chemistry studies using shock tube (ST) and rapid compression machine (RCM) facilities. Care must be used when interpreting and comparing autoignition data from ST and RCM studies. These facilities, which provide critical experimental knowledge of chemical kinetics, rely on buffer gases ranging from monatomics (typically Ar and He) to polyatomics (typically CO2) to control shock wave characteristics and thermodynamic conditions beyond those achievable with nitrogen alone. The composition of the buffer gases affects the specific heat capacity of the test gas mixtures, the three-body collision reactions, and the heat transfer rates from the test gas mixtures. Few studies consider these important and interrelated mechanisms; however, some researchers have identified concerns particularly with autoignition studies.
Hanson compared two reaction mechanisms for iso-octane and the predictions for autoignition times for $\phi = 1.0$, 2% $O_2$, 2 atm [6]. They considered argon and nitrogen as buffer gases, and found the change in specific heat capacity significantly affected the autoignition time (~50%) for one mechanism, but not the other, at 1400 K. At NUI-Galway, Wurmel et al. conducted an experimental and computational study of buffer gas composition (Ar, Xe, He, N$_2$) effects in an RCM for 2,3-dimethylpentane ($\phi = 1.0$, 15 atm, and 650-950 K) [7], and a computational study of iso-octane ($\phi = 1.0$, 2.0 atm) and methane ($\phi = 0.5$, 1.8 atm) each with Ar and N$_2$ buffer gases [8]. Wurmel et al. concluded that argon tends to decelerate autoignition times in their RCM and accelerate autoignition times in their ST, with different physical phenomena dominating the behavior over the wide timescale covered by RCM and ST facilities. Shen et al. conducted experimental shock tube studies of iso-octane over a range of equivalence ratios, pressures, and temperatures with argon and nitrogen as buffer gases, and made comparisons with three kinetic mechanisms [9]. Shen et al. determined that argon autoignition times were 20% shorter than nitrogen buffer gas experiments, primarily due to the lower heat capacity of argon, and that the mechanisms accurately captured the trend but generally failed to accurately predict autoignition times.

This study identifies using computational methods the effects of buffer gas composition across a broad range of thermodynamic conditions for three reference fuels: a branched alkane (iso-octane), a normal alkane (n-heptane), and an alcohol (n-butanol). Each of the fuels selected for this study have been previously investigated in the University of Michigan rapid compression facility [10, 11, 12, 13, 14], and the experiments provide the basis for the conditions studied. Two of the fuels selected are primary reference fuels and autoignition times have been extensively reported from other rapid compression machines (i-C$_4$H$_{10}$ [15, 16, 17, 18], n-C$_6$H$_{14}$ [19, 15, 18, 20]) and shock tubes (i-C$_4$H$_{10}$ [21, 22, 23, 24, 9, 25], n-C$_6$H$_{14}$ [21, 26, 27, 28, 29, 30, 31]) encompassing a wide range of conditions. Studies of n-butanol have occurred with greater frequency over the last 5 years, and autoignition data are available from another rapid compression machine [32] and shock tubes [33, 34, 35, 36, 37, 38, 39]. In this work, computational simulations of autoignition of test gas mixtures are used to quantify the thermal and kinetic effects of varying buffer gas composition and assign associated uncertainties on ignition delay time and heat release rate during autoignition. The study includes quantifying the effects of uncertainties in the species-specific collision efficiencies used in three body reactions.

### 2. Computational Methods

Computational simulations were carried out using the CHEMKIN suite of software (version 10113, x64) [40] and assuming a closed 0-D homogeneous batch reactor at adiabatic, constant volume conditions. Default values from CHEMKIN were used for the solver tolerances and solver time-steps. Detailed kinetic mechanisms were used and information on the mechanisms can be found in the literature for iso-octane [41], n-heptane [14], and n-butanol [34]. No modifications to reaction rates were made to the mechanisms considered in this study. The mechanisms used do not consider nitrogen (specifically NO, or HCN) chemistry. Initial conditions were selected based on relevance to internal combustion engine operating conditions and existing experimental data. The simulations were conducted at stoichiometric fuel-to-oxygen equivalence ratios ($\phi = 1.0$) over a temperature range of 600-1100 K in 25 K increments for all fuels. Dilution levels of 3.76:1 and 5.64:1 (inert gas to $O_2$ ratios, mole basis) were considered in this study. Argon, nitrogen, carbon dioxide, and water vapor were considered as buffer gases in this study. In order to identify the maximum impact of each buffer gas, one buffer gas was considered per mixture composition. Table 1 provides the initial conditions and mixture compositions studied.

Table 1. Initial conditions for 0-D homogeneous batch reactor simulations in CHEMKIN. Composition is provided on a mole basis.

<table>
<thead>
<tr>
<th>Fuel</th>
<th>$\chi$(fuel)</th>
<th>$\chi$(O$_2$)</th>
<th>$\chi$(diluent)</th>
<th>$\Phi$</th>
<th>diluent:O$_2$</th>
<th>$P_0$ [atm]</th>
<th>$T_0$ [K]</th>
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</thead>
<tbody>
<tr>
<td>i-C$<em>4$H$</em>{10}$</td>
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<td>20.66</td>
<td>77.69</td>
<td>1.0</td>
<td>3.76</td>
<td>9.0</td>
<td>600-1100</td>
</tr>
<tr>
<td>i-C$<em>4$H$</em>{10}$</td>
<td>1.91</td>
<td>14.88</td>
<td>83.93</td>
<td>1.0</td>
<td>5.64</td>
<td>9.0</td>
<td>600-1100</td>
</tr>
<tr>
<td>i-C$<em>6$H$</em>{14}$</td>
<td>1.91</td>
<td>14.88</td>
<td>83.93</td>
<td>1.0</td>
<td>5.64</td>
<td>60.0</td>
<td>600-1100</td>
</tr>
<tr>
<td>n-C$<em>6$H$</em>{14}$</td>
<td>1.87</td>
<td>20.61</td>
<td>77.52</td>
<td>1.0</td>
<td>3.76</td>
<td>9.0</td>
<td>600-1100</td>
</tr>
<tr>
<td>n-C$<em>6$H$</em>{14}$</td>
<td>1.34</td>
<td>14.90</td>
<td>83.76</td>
<td>1.0</td>
<td>5.62</td>
<td>9.0</td>
<td>600-1100</td>
</tr>
<tr>
<td>n-C$<em>6$H$</em>{14}$</td>
<td>1.34</td>
<td>14.90</td>
<td>83.76</td>
<td>1.0</td>
<td>5.62</td>
<td>60.0</td>
<td>600-1100</td>
</tr>
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<td>n-C$<em>6$H$</em>{14}$OH</td>
<td>3.38</td>
<td>20.30</td>
<td>76.32</td>
<td>1.0</td>
<td>3.76</td>
<td>3.2</td>
<td>600-1100</td>
</tr>
<tr>
<td>n-C$<em>6$H$</em>{14}$OH</td>
<td>2.45</td>
<td>14.71</td>
<td>82.84</td>
<td>1.0</td>
<td>5.63</td>
<td>3.2</td>
<td>600-1100</td>
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<tr>
<td>n-C$<em>6$H$</em>{14}$OH</td>
<td>2.45</td>
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<td>82.84</td>
<td>1.0</td>
<td>5.63</td>
<td>60.0</td>
<td>600-1100</td>
</tr>
</tbody>
</table>
3. Results and Discussion

Pressure-time histories were determined from CHEMKIN simulations for n-heptane, iso-octane, and n-butanol. Buffer gas composition was expected to alter the duration of the autoignition events and heat release rates. Ignition delay times for the simulations were defined as the time from the start of the simulation to the time corresponding to the maximum rate of pressure rise, dP/dt, as has been used previously [10, 11, 12, 13, 14]. Similarly, for conditions where two stage ignition occurs, the first stage ignition delay time is determined from the local maximum in the rate of pressure rise. The temperatures reported for the ignition delay times and pressure-time histories are the initial temperatures of the simulations. Figure 1 shows results for computed first stage ignition delay times (+ symbols) and the overall ignition delay times (dashed lines) in an Arrhenius diagram for the base case of n-heptane at φ = 1.0, P = 9.0 atm, diluent:O₂ = 5.62, over the temperature range 600-1100 K.

![Arrhenius diagram for computed ignition delay times of n-heptane. + symbols indicate the computed first stage ignition delay times. Initial conditions of P = 9.0 atm, χ(n-C₇H₁₆) = 1.34%, χ(O₂) = 14.90%, χ(diluents) = 83.76%.](image)

While only the results for n-heptane are presented in Figure 1, similar trends were observed for iso-octane at the same equivalence ratio, pressure, and dilution level. Within the negative temperature coefficient (NTC) region (~650 – 850 K, ~600 – 800 K for iso-octane), buffer gas composition has significant effect on the overall ignition delay time, with argon and water vapor accelerating times up to 31% (30% for iso-octane) and carbon dioxide decelerating times up to 49% (65% for iso-octane). However, the first stage of ignition shows little variance (< 7%, < 16% for iso-octane) across all of the buffer gases relative to nitrogen. Outside of the NTC region for both fuels, the buffer gases had less impact on the computed ignition delay times, with water vapor showing the most significant effect in the high temperature chemistry regime, accelerating times up to 37% (34% for iso-octane). In the high temperature chemistry regime for n-heptane (T > 900 K), argon and carbon dioxide did not alter ignition times more than 17% (15% for iso-octane).

For the baseline conditions of n-butanol at φ = 1.0, P = 3.2 atm, and diluent O₂ = 5.63, there was no NTC behavior over the range of temperatures considered, and the relative reactivity due to the buffer gases was consistent throughout the
temperature range. Water vapor and carbon dioxide accelerated ignition times up to 52% and 32%, respectively, compared to N₂, and argon decelerated ignition delay times less than 10% compared to N₂.

For the simulations conducted at high pressure (ϕ = 1.0, 60.0 atm, diluent:O₂ = 5.64), as expected the region of NTC shifted (e.g. to ~750 – 950 K for n-heptane, to ~700 – 900 K) compared to the low pressure simulations, and the general trends as a function of buffer gas were the same as observed at low pressure for the three fuels. However, the magnitudes of the buffer gas effects were diminished by ~20-50% for all fuels regarding the overall ignition delay times, with water vapor exhibiting the most significant reduction. Buffer gas effects on first stage ignition delay times also decreased ~20-50% for n-heptane, but changed less than 10% for iso-octane.

Simulations at air dilution levels for n-heptane, iso-octane, and n-butanol (ϕ = 1.0, 3.2/9.0 atm, diluent:O₂ = 3.76) showed similar trends across low and high temperature chemistry as the simulations at the more dilute 5.64:1 conditions for both low and high pressures. No significant shift in the NTC region (< 25 K) for n-heptane and iso-octane was achieved by decreasing the dilution level. The decreased dilution level of ~33% resulted in a ~35% decrease in ignition delay times outside of the NTC region for n-heptane/nitrogen and iso-octane/nitrogen mixtures. Within the NTC region n-heptane/nitrogen and iso-octane/nitrogen mixtures caused ignition delay times to decrease ~60% from the higher dilution results. Results from the n-heptane simulations are provided in Figure 2 along with experimental results at the same equivalence ratio and dilution level. Decreasing the dilution to air levels decreased the buffer gas compositional effects by ~30% in the NTC region and had minimal impact (< 10% change) on ignition delay times outside of the NTC region for all three fuels.

![Figure 2](image-url)

Figure 2. Arrhenius diagram for computed and experimental ignition delay times of stoichiometric mixtures of n-heptane at air dilution levels. + symbols indicate the computed first stage ignition delay times. Initial conditions of P = 9.0 atm, χ(n-C₇H₁₆) = 1.87%, χ(O₂) = 20.61%, χ(diluents) = 77.52%.

Solid orange symbols in Figure 2 represent experimental results from RCM and solid violet symbols denote ST results. The experimental ignition data were scaled using a 1/P factor to 9.0 atm across the entire temperature range considered.

For the equivalence ratio and dilution considered, Shen et al. [9] acquired data in the NTC region where the buffer gas
used was nitrogen alone. Data from the RCM studies required the use of three buffer gases (Ar, N₂, CO₂) to vary the end of compression temperatures over the range considered, so some scatter within the NTC region is expected due to buffer gas effects. The trends are in generally good agreement for the scaled experimental results and the computed ignition delay times. The experimental agreement is fair, within a factor of 2, outside of the strong NTC region (~700 – 850 K). However, in the strong NTC region the scaled experimental results vary significantly, spanning an order of magnitude in ignition delay times. This level of variation may have several causes including inappropriate scaling for pressure within the NTC region, to buffer gas composition effects, and differences in fundamental characteristics of the experimental facilities.

To separate the thermal effects from the kinetic effects of buffer gas composition on ignition delay times, the third-body collision efficiencies were set equal to 1, the collision efficiency of nitrogen, at conditions of φ = 1.0, P = 3.2/9.0, diluent:O₂ = 5.64. The resulting first stage and overall ignition delay times directly correlate with expectations based on specific heat capacity (i.e. τ₁Ar < τ₁N₂ < τ₁H₂O < τ₁CO₂) for all three fuels for all temperatures. The variance in first stage ignition delay times is < 7% for n-heptane (< 16% for iso-octane) across the buffer gases in the NTC region. Outside of the NTC region, buffer gases without enhanced (or reduced) collision efficiencies relative to N₂ changed the ignition delay by ±25%. Within the NTC region for n-heptane and iso-octane, the changes to the collision efficiencies changed the overall ignition delay time by nearly a factor of 2. The large difference in the sensitivity to buffer gas composition in the NTC region across thermodynamic conditions can be attributed largely to the role of specific heat capacity on two-stage ignition when collision efficiencies are compared at unity. Carbon dioxide and water vapor, possessing higher specific heats than nitrogen, exhibit smaller pressure and temperature increases during the first stage of ignition. Since mixtures with carbon dioxide and water vapor are significantly cooler after the first stage compared to nitrogen, the second stage of ignition is demonstrably longer for H₂O and CO₂ when collision efficiencies are set equal to 1. The opposite effect is true for argon when compared to a nitrogen buffer gas at unity collision efficiencies. When collision efficiencies are considered, as in Figure 1 and Figure 2 for n-heptane, carbon dioxide and water vapor move from decelerating ignition delay times in low temperatures (< 700 K for n-heptane at the baseline conditions) to accelerating ignition delay times (relative to nitrogen) as temperature increases for both pressures and dilution levels considered for both NTC fuels. Low-temperature chemistry shows little sensitivity to third-body collision efficiencies as the first stage ignition delay times are largely unaffected (< 3% change for each buffer gas) when they are set equal to 1. Specific heat capacities also have limited impact on low temperature chemistry as their effects across buffer gases are < 7% for n-heptane (< 16% for iso-octane) on first stage ignition delay times. Therefore ignition delay times for high temperature chemistry (> 850 K for n-heptane at the baseline conditions), to which the second stage of two stage ignition may be included, are significantly sensitive to third-body collision efficiencies which may produce effects equal to or greater than those from specific heat capacity differences for the conditions considered in this study. In the mechanisms considered, typical third-body collision efficiencies for carbon dioxide and water vapor are ~4 and ~10-20 times that of nitrogen respectively.

The effects of buffer gas composition on heat release rates were compared using a burn duration metric and using the time history of the pressure derivative. Based on the computed pressure-time histories, normalized 10-50% and 50-90% burn durations were calculated. Burn durations were normalized for each buffer gas using the 50% burn time for that gas at a given thermodynamic condition. Normalized 10-50% burn durations are provided in Figure 3 for n-heptane at φ = 1.0, P = 9.0 atm, diluent:O₂ = 5.62.
Figure 3. Results of sensitivity analysis for 10-50% burn duration normalized by the 50% burn time. Initial conditions of $P = 9.0 \text{ atm}$, $\chi(\text{n-C}_7\text{H}_{16}) = 1.34\%$, $\chi(\text{O}_2) = 14.90\%$, $\chi(\text{diluents}) = 83.76\%$.

Within the NTC region for n-heptane (and iso-octane) the 10-50% burn durations are longest for carbon dioxide accounting for as much as 77% (38% for iso-octane) of the ignition delay time. Such long burn durations in the NTC region are an artifact of the two-stage ignition. Outside of the NTC region, 10-50% burn durations remain below 20% (25% for iso-octane and n-butanol). All fuels exhibited the increase in 10-50% burn duration as temperature increased as seen in Figure 3 with carbon dioxide causing the longest normalized burn durations. Increased pressure and lower dilution levels both suppressed 10-50% normalized burn duration times for all three fuels.

Normalized 50-90% burn durations of n-heptane across all buffer gases remained under 5% for the range of temperatures and both dilution levels (3.76:1 and 5.64:1), with the largest values associated with CO$_2$ buffer gas. Elevated pressure (60.0 atm) caused the normalized 50-90% burn durations of n-heptane to increase significantly for CO$_2$, reaching 19% while the other buffer gases remained below 5%. Iso-octane and n-butanol displayed similar trends for normalized 50-90% burn durations across both dilution levels and elevated pressure, with durations remaining below 5% for all buffer gases with CO$_2$ causing the longest normalized burn durations.

Local sensitivity analysis of third-body collision efficiencies for the buffer gases were conducted for the three fuels at the baseline conditions ($\phi = 1.0$, $P = 3.2/9.0 \text{ atm}$, diluent:O$_2$ = 5.64). Third-body collision efficiencies were varied by a factor of 2 from their published values for the local sensitivity analysis. The local sensitivity was then defined as the difference between the new ignition delay time and the baseline ignition delay time, normalized by the baseline ignition delay time. Two reactions were found to have significant influence on the predicted ignition delay times and heat release rates,

$$H + O_2(\pm M) \rightleftharpoons HO_2(\pm M) \quad (R9)$$

$$H_2O_2(\pm M) \rightleftharpoons 2OH(\pm M) \quad (R18)$$
Local sensitivity results were symmetric for (R9) and (R18) for all fuels at the baseline conditions, i.e. doubling and halving collision efficiencies resulted in nearly the same magnitude of shift in the ignition delay time as seen in Figure 4 for n-heptane (ϕ = 1.0, P = 9.0 atm, diluent:O₂ = 5.62).

![Figure 4. Local sensitivity of ignition delay time to the collision efficiencies of the reaction, H₂O₂(+M)=2OH(+M). Initial conditions of P = 9 atm, χ(n-C₇H₁₆) = 1.34%, χ(O₂) = 14.90%, χ(CO₂) = 83.76%.

For all fuels, local sensitivities results for all the buffer gases remained below 30% for (R18) throughout the entire temperature regime at the baseline condition. Local sensitivity analysis of (R9) revealed that ignition delays were affected less than 10%/30%/5% for n-heptane/iso-octane/n-butanol respectively at the baseline conditions. The local sensitivities highlight the kinetic uncertainties associated with buffer gases and the effects on autoignition for the baseline conditions.

A pressure-time history and corresponding pressure rise rate from the local sensitivity analyses of stoichiometric n-heptane mixtures with carbon dioxide buffer gas at 9 atm, 5.62:1 dilution, and 700 K are shown in Figure 5.
Figure 5. Pressure-time history and heat release rates of n-heptane at low temperature (T = 700 K) conditions. Initial conditions of P = 9.0 atm, χ(n-C₇H₁₆) = 1.34%, χ(O₂) = 14.90%, χ(CO₂) = 83.76%.

Varying the collision efficiency of (R9) resulted in less than a 5% change of the ignition delay time, but significantly altered the pressure rise rate preceding the second ignition event for carbon dioxide. Enhancing the collision efficiency by a factor of 2 broadened the second ignition peak by ~20%, and reducing the rate by a factor of 2 narrowed the second ignition peak by ~20% for carbon dioxide. Local sensitivity of (R18) revealed no perceivable change (< 5%) in the duration of the either ignition peak for n-heptane for any buffer gas at the temperature presented. Water vapor showed similar effects to carbon dioxide on the pressure rise rate for n-heptane, while argon and nitrogen exhibited no such effects. Changes to the duration of pressure rise rates were not observed for iso-octane or n-butanol across any buffer gases for (R9) and (R18) at 700 K.

4. Conclusions

This study improves our understanding of the thermal and kinetic effects of buffer gas composition on the important combustion characteristics of ignition and heat release rate for three reference fuels. Simulations were presented for n-heptane, iso-octane, and n-butanol in stoichiometric mixtures with four buffer gases (Ar, N₂, H₂O, CO₂) for a range of pressures, dilution levels, and temperatures. Based on the conditions examined in this study, low pressures, high levels of dilution, and negative temperature coefficient conditions are most likely to result in significant thermal and kinetic effects due to the composition of buffer gases. Fuels that exhibit negative temperature coefficient behavior may show significantly (a factor of 2 or more) more pronounced buffer gas effects during two stage ignition. Caution should be exercised in the interpretation of data acquired in the NTC region from rapid compression machines and shock tubes when extrapolating the data from one buffer gas to another. The data are subject to potentially large uncertainties due to uncertainties in the collision efficiencies of the different buffer gases. Data collected at conditions outside the NTC region, or for fuels which do not exhibit NTC behavior, are unlikely to show significant (> 20%) effects of changes to buffer gas composition as < 30% variance was observed in this study for all fuels across both pressures and dilution
levels. Conversely, experiments to isolate the effects of buffer gas composition must have experimental uncertainties of less than $\pm 15\%$ to observe the changes predicted by these reaction mechanisms used in this study.

For fuels with similar combustion chemistry, the results of this study show that combustors are not likely see significant impact (> 20%) on autoignition chemistry and heat release rates when utilizing dilution techniques, such as exhaust gas recirculation, as significant effects were only observed using buffer gases of 100% CO$_2$ and 100% H$_2$O. Even for high EGR systems, N$_2$ is still over 50% of the buffer gas. However, as noted above, comparison of autoignition data across experimental facilities will be affected by buffer gas composition for studies focusing on negative temperature coefficient conditions.

There are two key limitations to this study. The results are linked to the global and local uncertainties of the coefficient conditions.

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