

Understanding the Chemical Effects of Increased Boost Pressure under HCCI Conditions

Emma J. Silke, William J. Pitz and Charles K. Westbrook

Lawrence Livermore National Laboratory

Magnus Sjöberg and John E. Dec

Sandia National Laboratories

ABSTRACT

One way to increase the load range in an HCCI engine is to increase boost pressure. In this modeling study, we investigate the effect of increased boost pressure on the fuel chemistry in an HCCI engine. Computed results of HCCI combustion are compared to experimental results in a HCCI engine. We examine the influence of boost pressure using a number of different detailed chemical kinetic models – representing both pure compounds (methylcyclohexane, cyclohexane, iso-octane and n-heptane) and multi-component models (primary reference fuel model and gasoline surrogate fuel model). We examine how the model predictions are altered by increased fueling, as well as reaction rate variation, and the inclusion of residuals in our calculations. In this study, we probe the low temperature chemistry (LTC) region and examine the chemistry responsible for the low-temperature heat release (LTHR) for wide ranges of intake boost pressure.

Results of this study are discussed, particularly focusing on the detailed calculations using the well-validated and chemically understood LLNL PRF mechanism. Inclusion of the effect of residuals improved the model's prediction of the onset and phasing of LTHR in PRF80 simulations. For the residual-containing simulations, the LTHR was advanced by the inclusion of residual to the fuel and air charge. This is due to the fact that by including residual in the calculation, the fuel and air charge contains reactive species (partial-burn products) from the onset of the calculation, and these species have the ability to advance the reactivity of the system.

INTRODUCTION

HCCI technology offers major advancement in high efficiency, low emission engines. This technology is highly desirable, since it combines the high fuel efficiency of Diesel or compression ignition engines with the less polluting abilities of spark-ignition engines. As in spark ignition engines, the charge is well mixed which

minimizes particulate emissions, and as in diesel engines, it is compression ignited and has no throttling losses, which all contribute to the high fuel efficiency [1].

In HCCI, the charge is diluted with excess air or exhaust gases to reduce the peak combustion temperature and consequently, NO_x emissions. Premixing of the fuel and air is usually achieved by injecting the fuel early in the intake stroke. HCCI engines can operate using a variety of fuels - gasoline, diesel, natural gas, bio-fuels and hydrogen. Ignition in the HCCI engine occurs nearly simultaneously throughout the volume when the homogeneous fuel and 'air' mixture are compressed to near top dead center (TDC). The ignition is controlled primarily by the chemical kinetics of ignition, with a secondary influence from effects such as turbulence and mixing [2].

Several technical difficulties with HCCI exist, and need to be resolved before HCCI can be implemented in production engines. Reducing the amount of unburned hydrocarbons and carbon monoxide emitted, expanding the load range of the engine operation and controlling the phasing of HCCI combustion are current problem areas. HCCI phasing is difficult to manage since it is managed by controlling the temperature, pressure, exhaust gas recirculation and composition of the fuel and air mixture so that it spontaneously ignites close to TDC where the maximum engine efficiency is realized. HCCI engines have been shown to operate well at low to medium loads, but obstacles have been encountered at high load conditions, at cold starts, and during transient operating conditions. Controlling ignition at all loads continues to be an issue, including the control of combustion phasing and limiting the heat-release rate (HRR) at high loads [3]. At high load conditions, avoiding too high of a pressure-rise rate so as to cause engine knock is one such problem.

One way to increase the load range is to increase the intake boost pressure. There are a number of challenges to increasing boost pressure in an HCCI engine. One of

the most significant is that the increased pressure enhances the autoignition reactivity of the fuel. Since overly advanced combustion phasing can substantially increase the pressure-rise rate and lead to engine knock [4], engine operating parameters must be adjusted to compensate for this effect. Controlling combustion phasing is particularly important under boosted conditions since the greater charge mass with boost increases the amount of pressure rise with combustion, which also increases the pressure-rise rate. Additionally, the increased autoignition reactivity can influence the burn duration. This is because at all but the lowest loads, the burn duration in an HCCI engine is controlled by thermal stratification. This causes the combustion to occur as a sequential autoignition of successively cooler zones [5-7], and this sequential autoignition will also be influenced by pressure boost. As a result, understanding the effect of intake-pressure boost on autoignition is critical for modeling boosted HCCI engines.

In the current work, we focus on understanding the controlling chemical kinetics of the autoignition of fuel-air charge under boosted operation in HCCI engines. We probe the low temperature chemistry (LTC) region under the conditions of increasing boost pressure. We have performed calculations for a number of pure fuel compounds, including n-heptane, iso-octane, methylcyclohexane, and cyclohexane, as well as a surrogate gasoline mixture and a primary reference fuel (PRF) mechanism. These calculated results are compared to experimental results from an HCCI research engine, previously described by Sjöberg and Dec [3,8-10]. In the next section, we further explore aspects of increased boost pressure.

INTAKE BOOST PRESSURE AND IGNITION-CHEMISTRY REGIMES

Figure 1 shows the effect of increasing the absolute intake pressure from 80 to 180 kPa for PRF80 (primary reference fuel) using the LLNL detailed chemical kinetic model [11]. As the intake (or boost) pressure is increased, the temperature at bottom dead center (BDC) must be reduced to maintain the phasing of the heat release near TDC to achieve the efficiency gains desired with HCCI engines and to prevent knocking. At the high boost pressure condition, the fuel/air mixture attains lower temperatures and higher pressures compared to the unboosted 80kPa case. This can be clearly seen by comparing, for example, Figs. 1(b) and 1(d). These boosted conditions enable the onset of low temperature reactions and an accompanying temperature increase seen at 350°CA (crank angle degrees).

The combined change of intake pressure and temperature results in the fuel and air mixture traversing different temperature and pressure regimes during the autoignition process. Mehl *et al.* [12] allude to this effect in their recent paper using detailed chemistry to understand both octane numbers and gasoline sensitivity. They report their findings using detailed pressure-temperature-time topographical reactivity maps

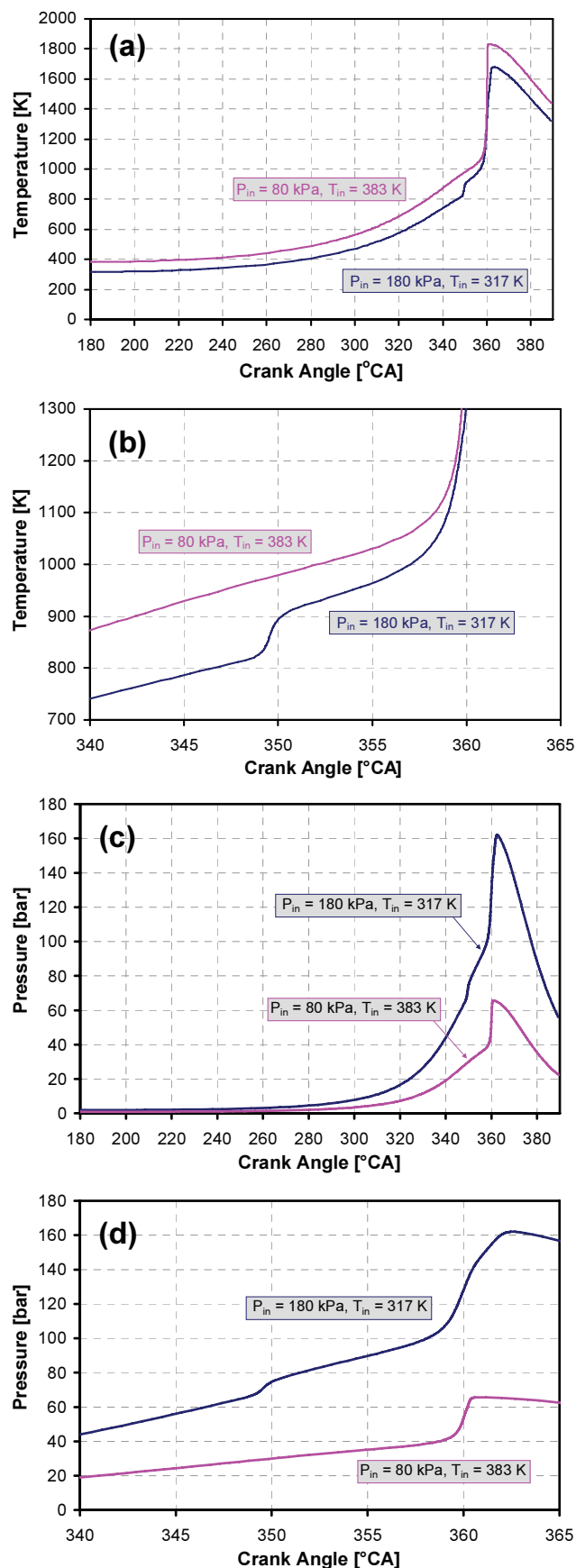


Figure 1. Simulations for PRF80 (80% iso-octane and 20% n-heptane), $\phi = 0.28$ and two different boost levels. (a) - temperature history, (b) - zoomed-in temperature, (c) - pressure history, (d) - zoomed-in pressure.

and show that for two different fuels (n-pentane and 1-pentene), increasing the pressure forces the fuel to different chemistry regimes at different time intervals.

The effect of increased boost pressure, illustrated in Fig. 1, affects both the occurrence of low-temperature heat release (LTHR) and the onset of high-temperature heat release. Hwang *et al.* [13], and Sjöberg and Dec [14,15] have discussed the combined effects of LTHR and intermediate-temperature heat release (ITHR) both contributing to the observed onset of hot-ignition combustion. They found that in addition to the LTHR, PRF80 has more active ITHR than PRF100. This leads to a lower temperature requirement for the onset of high-temperature heat release for PRF80. This example implies that a chemical-kinetics model needs to predict both the correct amount of LTHR, as well as the correct ITHR to obtain the correct temperature and phasing for the onset of hot-ignition combustion.

One of the methods used to control the combustion phasing and HRR is through the ignition properties of the fuel in use. Two-stage ignition fuels have been shown to offer significant advantages for controlling combustion phasing and extending to high-load operation [15]. These fuels have an early LTHR which is associated with the occurrence of the low temperature chemistry (LTC) event. Such heat release causes an increase in the temperature of the charge and accelerates the reactions leading to the main ignition event (see Fig. 1b for one example). As a result, to maintain combustion phasing, the intake temperature (T_{in}) must be reduced by an amount proportional to the amount of LTHR [3]. Experiments by Sjöberg and Dec have shown that the amount of LTHR increases with the fuel/air equivalence ratio (ϕ), [10,17]. As such, the ignition timing will be influenced by the local fuel concentration. Consequently, two-stage fuels offer potential for combustion-phasing control by fuel stratification created by delayed fuel injection [17].

Since the LTHR of two-stage fuels reduces the required T_{in} and thus increases the intake charge density, two stage fuels provide improved power density for typical HCCI engine configurations. Control of the amount of LTHR is significant since it affects the timing of the hot ignition event. As alluded to earlier in this discussion, this heat release is a property of the fuel in use, and its molecular structure, and it will also be influenced by the physical operating conditions in use, including temperature, pressure, and equivalence ratio.

Increased boost pressure can be used to increase autoignition reactivity. Naik *et al.* [18], Christensen *et al.* [19] and Westbrook *et al.* [20] have all shown this to be the case for fuels including gasoline, methylcyclohexane, iso-octane and n-pentane. In the same recent publication of Sjöberg and Dec [3] discussed earlier in this paper, they examined whether intake pressure adjustment could be used effectively to control the LTHR, using a constant engine speed of 1500 rpm and a blend of 30% n-heptane, 40% toluene and 30% iso-

octane. They report the effect of intake pressure or P_{in} on the T_{in} requirement in order to maintain the 50% burn point, and showed that the effect of intake boost pressure is strong, requiring that the T_{in} drop rapidly with increasing P_{in} . Any increase in P_{in} , in the pressure, temperature and fuel regime studied, causes the amount of LTHR to increase rapidly, thus the need for T_{in} to be reduced as P_{in} increases. The objective of the present work is to investigate the changes in the underlying chemistry as the boost pressure is increased in an HCCI engine. This investigation is conducted using chemical kinetic models for a number of different practical fuels. Further understanding the underlying chemistry may help to determine how to increase the boost pressure range for HCCI operation.

MECHANISM AND MODEL DETAILS

The effect of boost pressure on HCCI combustion was explored using the CHEMKIN-III suite of codes [21] and various detailed chemical kinetics mechanisms. All of the mechanisms used are those previously published by Lawrence Livermore National Laboratory, the details of which are as follows: n-heptane [16], iso-octane [22], methylcyclohexane [23], cyclohexane [24], gasoline surrogate mechanism [18] and PRF mechanism [11].

For the numerical model, we used the Senkin application which is a single-zone model that treats the in-cylinder charge as a single lumped mass with uniform composition and thermodynamic properties. The model allows the charge to be compressed and expanded adiabatically using the standard slider-crank relationship. The particular version of Senkin we use has a modified post-processor, as described by Dec and Sjöberg [8], where the heat release rate, total hydrocarbon (HC) species and total oxygenated hydrocarbon species (OHC) at each time step, as well as the standard output, may be extracted from a simulation run. This is especially useful in this work, since comparing the experimental LTHR against the model-predicted LTHR gives us an indication of how well we are able to simulate the engine event, and the ability to evaluate the differences in low temperature reactivity.

In general, the single-zone model is extremely useful in that it allows investigation of chemical kinetics and observations to be made about how these influence in-cylinder processes fundamental to HCCI combustion. In addition, the single-zone model allows for full chemical kinetic mechanism to be used for the fuel under study. Indeed, multi-component surrogate fuel models representative of gasoline fuel can be used, with relatively short computational times required compared to multi-zone calculations or computational fluid dynamic (CFD) calculations with reacting flows. This facilitates the investigation of ignition characteristics of different fuels under HCCI conditions, gaining insight into the chemical kinetics controlling the ignition event. The single-zone model treats the entire charge as a uniform entity, which is very much an oversimplification of a real engine environment where the charge will never be

completely homogeneous, since boundary layers, crevices, and in-cylinder convective transport will cause thermal non-uniformities even if the fuel, air and residuals from the previous cycle are well mixed. However, the model should still provide a good representation of typical fuel/air parcels within the central part of the charge, and therefore, it should be useful for examining trends in the ignition behavior of the bulk gases. We explored the effect of increasing boost pressure in an HCCI engine on the chemistry of various fuels and fuel mixtures. We started by examining our most recent chemical-kinetic fuel mechanisms under HCCI boosted conditions and ended by examining our most well validated and well-established fuel models. Finally, we examine the effect of including the residual fraction from the previous cycle on the calculated results.

CYCLOALKANES

Most recently we published a detailed chemical kinetic mechanism for the oxidation of cyclohexane (CHX), the simplest cycloalkane [24]. This work was a follow-up study to the detailed kinetic mechanism assembled by Pitz *et al.* [23] for methylcyclohexane (MCH) oxidation. In study of MCH autoignition, it was shown that when estimating the low temperature reaction rate rules for cycloalkane compounds that cyclic hydrocarbon systems are significantly different from acyclic compounds and this must be considered when determining rate rules. For example, the rate constants of alkylperoxy isomerisations are quite different for a cyclic molecule versus an acyclic molecule. Our more recent work on cyclohexane was motivated by the need to address such differences, and we did so by examining the case of the simplest cycloalkane, cyclohexane (CHX) [24]. This allowed fundamental reaction rate rules, including the similarities and differences in the correction terms between cyclic and acyclic isomerisation reactions, to be determined and well understood.

We have used our models to simulate the behaviour of cycloalkanes in a HCCI engine with variation in boost pressure. First in a previous publication [18], the methylcyclohexane (MCH) experiments of Sjöberg and Dec were modelled (Fig. 2). These results are discussed below. In the experiments, the combustion phasing, as measured by the 50% heat release point (CA50), is always kept at TDC to maximize the engine efficiency. As the boost pressure is increased, the bottom-dead-center (BDC) temperature must be reduced to maintain CA50 phasing. For MCH, the decrease in BDC temperature is especially dramatic at high boost pressure. This effect is due to the onset of low temperature heat release observed in the experiments. However, the MCH model did not capture the behaviour seen at high boost pressure where BDC temperature drops off dramatically.

In our current work, we used our most up-to-date cycloalkane chemistry model (for CHX) in an effort to see if using this we could get the high boost pressure,

reduced intake temperature fall-off that is observed in the experiments for methylcyclohexane.

Figure 2 shows the results for cyclohexane, and in this instance, the simulations did not manage to capture the high boost pressure, temperature fall-off either.

We then increased the equivalence ratio (ϕ , ϕ) – or increased the ‘fueling rate’ – in order to see if the fall-off could be captured by our model by doing so, Fig. 3. Experiments have shown that increased fueling causes more low temperature chemistry to occur [8,9,17]. The equivalence ratio was varied from 0.2 to 0.5, in increments of 0.1.

Figure 3 clearly show that at increased fueling, the model did not show a temperature fall off at high boost pressure, as was observed in the experiments for methylcyclohexane. In addition, the model shows less slope at lower pressure where low temperature heat release is not seen in the experiments. There is however, an observable difference as ϕ is varied from 0.2 to 0.5, such that as the fuel concentration is increased, the model predicts that a higher TDC temperature is required, and the slope of the line increases slightly. This is probably due to the effect of increased fuel on the specific heat and thus gamma (γ)

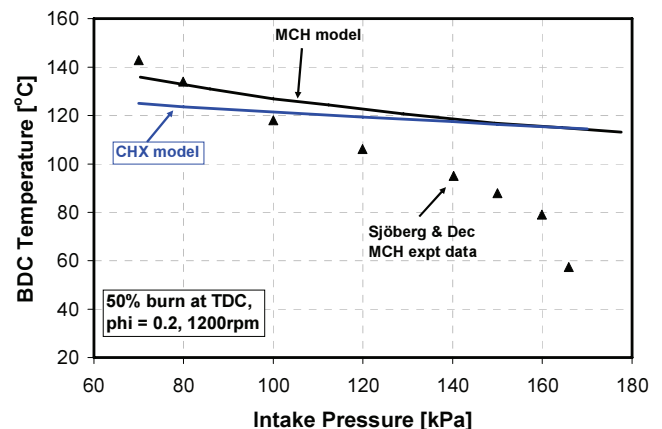


Figure 2. Simulations of MCH, CHX, compared to experimental data for MCH.

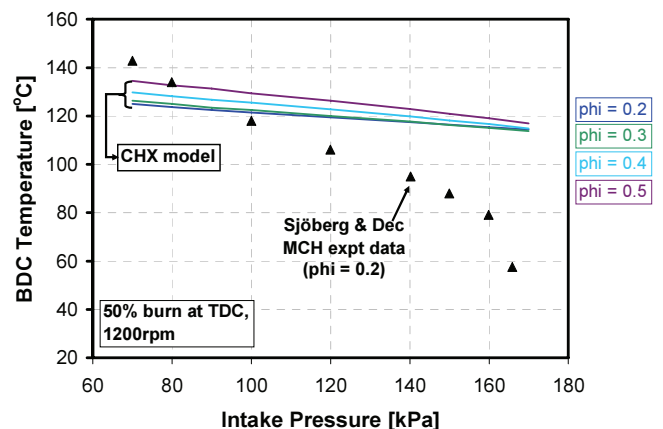


Figure 3. Simulations showing variation in the fueling rates for CHX calculations.

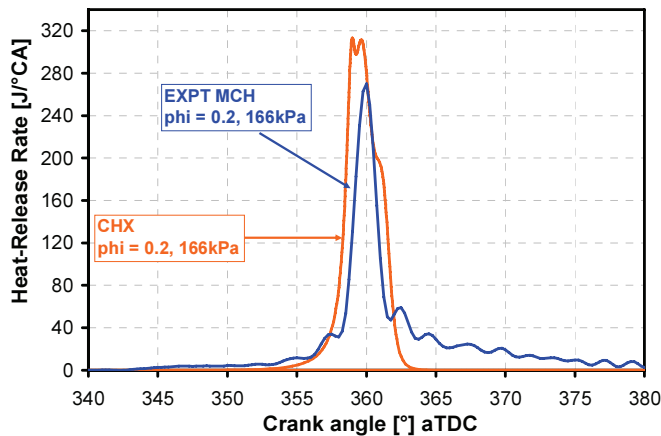


Figure 4. Heat release profiles for MCH (experimental) and CHX (simulation).

of the mixture leading to a decreased temperature at TDC. The same effect on the compressed gas temperature is seen when the γ of gas is changed by varying the diluent gas composition of a reacting mixture [25].

In the case of the CHX simulations, we also examined the HR profiles for these runs against the experimentally measured HR profiles for MCH – a sample comparison is made in Fig. 4. The HR profiles in HCCI engines can be divided into three stages: 1) the onset of low temperature chemistry, 2) high temperature heat release (or hot ignition) and finally 3) the conversion of CO to CO₂. These individual stages are not particularly apparent in the experimental HR profile MCH - stage 1 is not especially distinct, only that the experimental line increases slightly between 343 and 355°CA. Stage 2 is definite for both the experiment and the model (CHX). The model appears to capture two of the defined regions well, stage 2 and 3. To further investigate the effect of boost pressure, we next compare results of our gasoline surrogate model with experimental data on HCCI as a function of boost pressure.

GASOLINE

We now examine the behavior of gasoline under HCCI conditions with increased boost pressure. Since conventional fuels, like gasoline, consist of hundreds of components, it is not feasible to incorporate all of these chemical compounds into the development of a gasoline kinetic model. Instead, for surrogate fuel model development, various fuel components are selected and included, to represent the major chemical classes found in a typical gasoline. This results in a surrogate mechanism containing a representative component for each molecular structure of the various chemical classes found in a typical gasoline fuel. These include n-paraffins, iso-paraffins, olefins, cycloalkanes and aromatic compounds. This strategy has previously been reported by Naik *et al.* [18]. Generally, the composition of the surrogate model fuel is dependent on the target properties that are required to be predicted, (e.g. ignition behavior), so the nature of the various chemical classes

TABLE 1. Molar composition of the gasoline surrogate fuel mixture used in this work.

% Molar Composition	Mixture 1
iso-Octane	60
n-Heptane	8
Toluene	20
Methylcyclohexane / Cyclohexane	8
1-Pentene	4

represented and the amount of the representative components may be adjusted accordingly.

The gasoline surrogate mixture that we considered uses n-heptane to represent the n-alkane class, iso-octane to represent the iso-alkane class, 1-pentene to represent the olefin class, methylcyclohexane to represent the cycloalkane class, and toluene to represent the aromatic class. This is the same mixture of surrogate components as chosen by Naik *et al.* in their Mixture 1 [18]. Table 1 summarizes the proportions of each of these representatives of the chemical classes used in the surrogate fuel model. The difference in this current work is that we changed the cycloalkane component from methylcyclohexane to cyclohexane, so that we can utilize our most recent chemistry model to represent the cycloalkane class of compounds.

Figure 5 depicts the results obtained for simulations using the gasoline surrogate mechanism with cyclohexane, as well as showing results from the surrogate mechanism with methylcyclohexane. Indeed, there is very little difference between the two sets of calculations, only in that the version with cyclohexane is igniting marginally faster or requires a lower BDC temperature than that calculated with methylcyclohexane as the representative cycloalkane. Most importantly, the fall off behavior exhibited by the experimental data at high boost pressure and low intake temperature is not captured by either version of the gasoline surrogate fuel model.

As with the pure cyclohexane simulations, we increased the fueling rate (or ϕ) in the simulation to see if this resulted in a fall-off at high boost pressure. The fueling rate was varied from equivalence ratios of 0.2 to 0.5, in increments of 0.1, and the results are shown in Fig. 6. Unlike the simulation results obtained with pure cyclohexane, increasing the fueling of the gasoline surrogate fuel model was seen to have a significant effect on the model predictions.

Indeed as the fuel was increased, the distinct fall off was captured by the model, with a fueling rate of 0.4 most closely resembling the experimental data, as shown in Fig. 6. Additionally, the HRR profile of the model-simulated result at $\phi = 0.4$ and 190 KPa matched the experimental HR profile at $\phi = 0.2$ and 166 KPa well up to 358° CA, Fig. 7. This figure shows similar