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Authors

Chen, Yi-Hann
Chen, J Y

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Development of Isooctane Skeletal Mechanisms for Fast and Accurate Predictions of SOC and Emissions of HCCI Engines based on LLNL Detailed Mechanism

Y.-H. Chen and J.-Y. Chen
Department of Mechanical Engineering
University of California at Berkeley
Berkeley, CA 94720

Abstract: Fast and accurate numerical analysis is not only important for studying Homogeneous Charge Compression Ignition (HCCI) technology but also critical for designing HCCI engines. Chemistry plays the major role in determining Start of Combustion (SOC) and emissions of HCCI engines. The Lawrence Livermore National Laboratory (LLNL) detailed isooctane mechanism contains 857 species and 3,606 reaction steps making the calculation too expensive. This work describes a recent development of isooctane skeletal mechanisms for speeding up numerical simulations of HCCI. By using the rate analysis, two skeletal mechanisms were constructed: one with 258 species and the other with 291 species. The former was developed for accurate predictions of SOC and the latter is an expanded version of the one with 258 species aiming at accurate predictions of both SOC and emissions. Validations of the performances of these two skeletal mechanisms were conducted extensively for the operation regimes anticipated by HCCI engine applications. Both skeletal mechanisms are found satisfactory in predicting SOC with a speeding up factor of 15-20. The expanded version is found necessary for accurate predictions of CO and unburned hydrocarbon emissions.

1 Introduction

Numerical simulations have become an important tool for enhancing our understanding of combustion processes in internal combustion engines. The Homogeneous Charge Compression Ignition (HCCI) process has the fortunate characteristic of being relatively insensitive to turbulence and mixing effects, compared to spark ignition or compression ignition engines. Chemistry is found to be the main parameter in determining the Start of Combustion (SOC). An efficient and accurate numerical analysis for HCCI needs to include an adequate description of combustion chemistry. Recent advancements in detailed descriptions of combustion processes have led to a drastic increase in the total number of chemical species and steps in the detailed

chemical mechanisms. For gasoline engine applications, the detailed isooctane mechanism developed by Lawrence Livermore National Laboratory (LLNL) contains 857 species and 3,606 steps (Curran et al, 2002). Although the homogeneous feature of HCCI allows us to simplify the treatment of fluid dynamics by using a Well-Mixed Reactor (WMR) (e.g., Flower et al, 2000), a typical run with the detailed LLNL mechanism takes about half hour for one HCCI cycle on an Intel Pentium 2.8 GHz processor. More advanced models, such as the multi-zone model, would require many hours to perform. In order to significantly cut down the computational time, a reduced description of combustion chemistry with fewer species and steps can be used (e.g., Aceves et al, 2002). In addition, a reduced description can be used for speedy training of artificial neural networks for emissions or ignition delay (Traver et al 1999; Choi and Chen, 2005). This paper describes the development and validation of two skeletal mechanisms with accurate predictions of SOC and emissions. The paper is organized as following: first, a brief description of the methodology in developing skeletal mechanisms is given. Next, an extensive evaluation of the performances of the skeletal mechanisms is presented and followed by concluding remarks.

2 Development of Skeletal Mechanisms

Skeletal mechanisms are developed by evaluating the importance of each species and reaction step for the chemical processes of specific regimes of interest. Unimportant reaction species and steps are judiciously removed. As such, skeletal mechanisms are truncated versions of detailed mechanisms and they can be used only for the targeted regimes. Typically, numerical data are collected from simulations with detailed mechanisms performed for the targeted regimes of interest. With the numerical data, several methods can be used for developing skeletal mechanisms including rate analysis (e.g., Wang and Frenklach, 1991), sensitivity analysis (e.g., Yetter et al, 1991), and Computer Singular Perturbation (CSP) (Lam, 1993). As chemical kinetics is highly nonlinear and dependent on the regimes of interest, each approach has its own advantages and shortcomings. In principle, combinations of these methods could be used for developing skeletal mechanisms but such a comprehensive approach has not been fully developed yet. Alternatively, an empirical approach has been developed by using generic algorithms to optimize the rate constants for the reaction steps retained in the skeletal mechanisms (Patel et al 2004). Due to the empirical nature of such an approach, the applicable regime may be very limited.

When a large detailed mechanism is considered, the computational effort to generate the numerical data for the targeted regimes becomes an influencing factor in choosing the method for developing skeletal mechanisms. Both sensitivity analysis and CSP require information on the Jacobian matrix, say $\partial T / \partial k_i$, where k_i is the reaction rate of i -th step. Such information is expensive to generate when the detailed mechanism contains a large number of species and steps. For instance, using a single-zone Well Mixed Reactor (WMR), Aceves et al (2002) computed the sensitivity information with the detailed LLNL mechanism for the purpose of developing skeletal mechanisms. A typical run time took about 10 hours for each HCCI cycle on an Intel Pentium 2.8 GHz PC. Therefore only a handful of numerical data were generated. Although a reasonably accurate skeletal mechanism containing 197 species was developed, its usage is limited to prediction of SOC for a narrow engine operation regime, and emissions of carbon monoxide (CO) and unburned hydrocarbons (HC) were not targeted.

In order to construct skeletal mechanisms rapidly, we found it possible to develop reasonably accurate skeletal mechanisms with the rate analysis that requires the least computer time in generating numerical data. In the present study, numerical data were generated with the detailed LLNL mechanism using the single-zone WMR for the conditions listed in Table 1. These sample conditions were chosen to be representative of expected HCCI gasoline engine operations. Engine wall temperature was kept at 157°C (430K) and the overall heat transfer of reactive gas to the cylinder walls was modeled using the Woschni's correlation (e.g., Heywood, 1989). Due to the large size of the detailed mechanism as well as many trials needed before reaching satisfactory results, skeletal mechanisms were constructed with the assistance of an automatic computer program (Chen, 2001). For each engine condition, several records of numerical data were collected at -5, 0, 5, and 10 Crank Angle Degrees (CAD) relative to the Top Dead Center (TDC). This series of data intends to cover the most active chemical states just before and after SOC. All these data were input to the automatic computer program for identifying the importance of each reaction step. The rate analysis uses both reaction rate and heat release as criteria for importance. A reaction step is included in the skeletal mechanism when its normalized reaction rate or heat release rate exceeds preset limits. More specifically, a reaction step is considered important when its forward or reverse rate normalized by the maximum reaction rate exceeds a limit, RL . Similarly, when the normalized heat release rate of a reaction step exceeds a threshold, HL , this step is included in the skeletal mechanism. These two limits (RL and HL) were adjusted by numerical experiments with different trial and error combinations of RL and HL values until acceptable results were obtained. Such numerical experiments were practical only with the

assistance of an automatic computer program. For the present HCCI cases, satisfactory skeletal mechanisms were developed with $RL=10^{-6}$ and $HL=2 \times 10^{-6}$ after several trials manually. In the future, it may be possible to optimize the combination of RL and HL by optimization procedures using genetic algorithms (e.g., Gen and Cheng, 2000), or Integer Program (Bhattacharjee et al, 2003).

Based on the above rate analysis, two skeletal mechanisms were developed with satisfactory results: one with 258 species and 621 steps, referred to as Skeletal258, and the other one with 291 species and 875 steps, referred to as Skeletal291. Skeletal258 was developed from numerical data obtained for equivalence ratio (ϕ) =0.6 and inlet pressure at 2 atm. Skeletal258 was found to give accurate predictions of SOC over a wide range of conditions while keeping the total number of species as small as possible. During the course of development, several trial skeletal mechanisms were constructed with other engine conditions but the above specific condition yields the best results with the least number of species. Again, using computer optimization procedures could reduce human effort in this part of development.

Since accurate predictions of CO and HC are equally important for proper designs of HCCI engines, Skeletal291 was developed from results obtained for both high ($\phi=0.6$) and low equivalence ratios ($\phi=0.2$). The low equivalence ratio run contains high level of CO and HC, hence bringing in extra species and steps important to emissions. As such, Skeletal291 can be considered as an expanded version of Skeletal258 for accurate predictions of both SOC and emissions at the expense of additional 33 species.

3 Validation of Skeletal Mechanisms

A comprehensive evaluation of the performances of the skeletal mechanisms was conducted to define their applicable regimes. For HCCI applications, three different categories were evaluated: 1) ignition delay times (τ), 2) transient multi-cycle engine simulations with WMR, and 3) CO and HC emissions under very lean conditions. The first two examine model predictions of SOC and the last one focuses on emissions at idle conditions.

Ignition delay times:

For the first category, a wide range of conditions has been tested as listed in Table 2. A total of 36,750 runs were conducted using both the detailed LLNL and the two skeletal mechanisms with Chemkin II (Kee et al, 1992) for a constant pressure reactor. With the detailed mechanism, each run on the average takes about 10 minutes on an Intel Pentium 2.8 GHz PC. Two computers were used to perform runs on these conditions over a period of about four months. The average CPU time per run is cut to about 0.5-0.7 minutes when the skeletal mechanisms are used; a speeding up factor of 15-20 is realized. On the average Skeletal291 used more CPU time than Skeletal258. Although Skeletal291 contains more species than Skeletal258, under certain lean conditions, the CPU time with Skeletal291 is found shorter than that with Skeletal258. A plausible explanation is that since some important intermediate reaction steps and species are absent in Skeletal258, the chemical system becomes stiffer under certain conditions causing longer CPU time to compute.

Figure 1 presents example comparisons of predicted ignition delay times versus $1000/T$, where T is the initial mixture temperature in Kelvin. The delay times were computed for a constant-pressure reactor at three pressures ($P=10, 20, \text{ and } 40 \text{ atm}$) and two equivalence ratios ($\phi=0.3 \text{ and } 0.6$). As seen in the figure, both skeletal mechanisms give results almost identical to those obtained with the LLNL detailed mechanism when the temperature is higher than 1000K ($1000/T < 1$). Compared to the LLNL mechanism, both skeletal mechanisms yield longer ignition delays when the initial temperature is lower than 1000K. For HCCI engine applications, the reactive gas temperature near the TDC usually reaches 1000K or higher in order to avoid misfires. Therefore, accurate predictions of ignition delays at temperatures higher than 1000K are more critical than low temperatures.

Using the ignition delays, we can roughly estimate the errors in SOC predictions by using the skeletal mechanisms in terms of CAD. For instance, we assume that there is no chemical reaction until the pressure and temperature are 40 atm and 1000K respectively inside an HCCI engine. Using the differences in the predicted ignition delays between a skeletal mechanism and the detailed mechanism, we can translate the differences in ignition delays into differences in SOC predictions in terms of CAD at a certain engine speed, say 1800 RPM. The transformation between the difference in ignition delays and the difference in predicted values of SOC in terms of CAD is given by

$$\Delta CAD(1800) = \Delta(\tau [sec]) \cdot \frac{1800 [RPM]}{60 [sec/min]} \cdot 360 [CAD / revolution], \quad (1)$$

where $\Delta(\tau)$ = ignition delay by LLNL detailed mechanism – ignition delay by skeletal mechanism. For other engine speeds, the difference is linearly scaled with engine speed as ΔCAD (engine speed) = ΔCAD (1800) x engine speed/1800. Note that $\Delta CAD(1800)=1$ corresponds to $\Delta(\tau)=9.25 \times 10^{-5}$ s. Figure 2 presents an example comparison of ΔCAD (1800) versus $1000/T$ at 40 atm and several equivalence ratios for the two skeletal mechanisms. As observed in Figure 2(a), ΔCAD (1800) of Skeletal291 is less than 0.4 CAD for temperatures at or above 1000K and the accuracy deteriorates at lower temperatures. In comparison to Skeletal291, Figure 2(b) shows that for temperatures greater than 1000K, ΔCAD (1800) of Skeletal258 is less than 0.2 CAD except for $\phi=0.3$ at which maximum ΔCAD (1800) is 3 CAD. This trend is not surprising as Skeletal258 was developed from conditions at $\phi=0.6$. Comparisons of ΔCAD at other conditions all exhibit similar trends and accuracy (not shown due to limited space); therefore, it is concluded that the two skeletal mechanisms provide good predictions of SOC over the conditions listed in Table 2.

Transient Multi-cycle Engine Simulations with WMR

Simulations with the transient WMR model over several cycles can provide a more realistic evaluation for the performances of skeletal mechanisms under HCCI operations. In addition, the effect of EGR on SOC can be better assessed over multiple cycles. To keep the simulation simple, the intake and exhaust processes are not included in the calculations. Figure 3 shows two example comparisons of results obtained with the CFR engine parameters (Table 1) except different values for intake equivalence ratio, mixture temperature at the Bottom Dead Center (BDC), and EGR. In Figure 3(a), the first cycle was run with $\phi=0.3$ without EGR. In the second and third cycles, EGR is set to 0.3. In the simulation, the equivalence ratio is referred to the mixture in the intake charge. In the second cycle, the in-cylinder charge at the BDC is computed as a well-mixed mixture of fresh charge and the exhaust gas according to the amount of EGR. The temperature at BDC is set to 150°C for all the cycles. Successful HCCI combustion cycles are computed and the results from both skeletal mechanisms are in excellent agreement with those from the LLNL detailed mechanism. In Figure 3(b), the first cycle was run with the intake charge equivalence ratio set to 0.6 without EGR. In the following cycles, the intake charge equivalence ratio is set to 1 with EGR=0.6. The temperature at BDC is set to 140°C for the first cycle and 150°C for the following cycles. Under this particular set of conditions, the computed results exhibit alternative burning and misfire cycles after third cycle. This type of engine

behaviors is extreme and not meant for practical usage. This comparison merely shows that the two skeletal mechanisms are capable of reproducing chemical kinetics under extreme conditions.

CO and Unburned Hydrocarbon Emissions

Emissions of CO and HC are important issues in HCCI engines. Most emissions are resulted from engine crevice areas. In HCCI gasoline engine applications, running at very lean conditions is considered as a potential means for achieving idle without a throttle plate. When the equivalence ratio decreases, CO and HC in the bulk gas phase are expected to increase. The performances of skeletal mechanisms in predicting emissions of CO, HC, and oxygenated unburned hydrocarbons (HCO) are assessed by running the single-zone WMR using the engine parameters listed in Table 1 with (EGR=0.3) and without EGR for equivalence ratios ranging from 0.075 to 0.25. Figure 4 compares the predicted CO emissions from the bulk gas phase with/without EGR. CO emissions are seen to increase with equivalence ratio, reaching the peak and then decrease with ϕ . Addition of EGR shifts the peak to a richer mixture. Comparisons reveal that Skeletal291 yields results in excellent agreement with those obtained with the detailed LLNL mechanism. Skeletal258 gives results that capture the trend but much less accurate.

The corresponding comparisons of predicted HC and HCO are presented in Figure 5, both showing a decrease trend with equivalence ratio, ϕ . Clearly, the predictions by Skeletal258 contain large errors in both HC and HCO, and even show incorrect trends for HCO. In contrast, Skeletal291 gives results following faithfully the results by the detailed LLNL mechanism.

4 Conclusion

Based on the detailed LLNL isooctane mechanism, two skeletal mechanisms, denoted as skeletal258 and skeletal291, have been developed and validated for HCCI applications. Overall, both skeletal mechanisms predict the start of combustion in good agreement with those from LLNL detailed mechanism over a wide range of conditions. Skeletal291 contains 33 more species than skeletal258. Through extensive evaluations, Skeletal291 is found necessary for accurate predictions of CO and HC emissions. A speed-up factor of 15-20 has been realized using the skeletal mechanisms. Although the skeletal mechanisms may be still too large for calculations using three-dimensional computational fluid dynamics, the skeletal mechanisms can be readily

used in single-zone or multi-zone HCCI models for reasonably accurate predictions with much less computing time than with the LLNL detailed mechanism.

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Table 1 Engine specification and parameters used by WMR

CFR Engine	Value	Operation parameters	Value
Displacement (liter)	0.612	Inlet temperature (°C)	150, 160
Number of cylinder	1	Inlet pressure (atm)	1, 2
Bore (mm)	82.5	Equivalence ratio	0.2, 0.3, 0.6
Stroke (mm)	114.	EGR (%mass)	0, 30, 60
Connecting rod length (mm)	254.	Engine speed (rpm)	1,800
Compression ratio	16.	Fuel	Isooctane

Table 2 Engine parameters for evaluations of ignition delays

Parameters	Conditions	Total number of points
Equivalence Ratio	0.3, 0.45, 0.6, 0.8, 1.0	5
Pressure (atm)	5 ~ 100 (equally spaced with $\Delta P=5$ atm) 100 ~ 200 (equally spaced with $\Delta P=10$ atm)	30
Temperature (Kelvin)	600 ~ 1800	49
Exhaust Gas Recirculation (EGR)	0.0, 0.2, 0.4, 0.6, 0.9	5

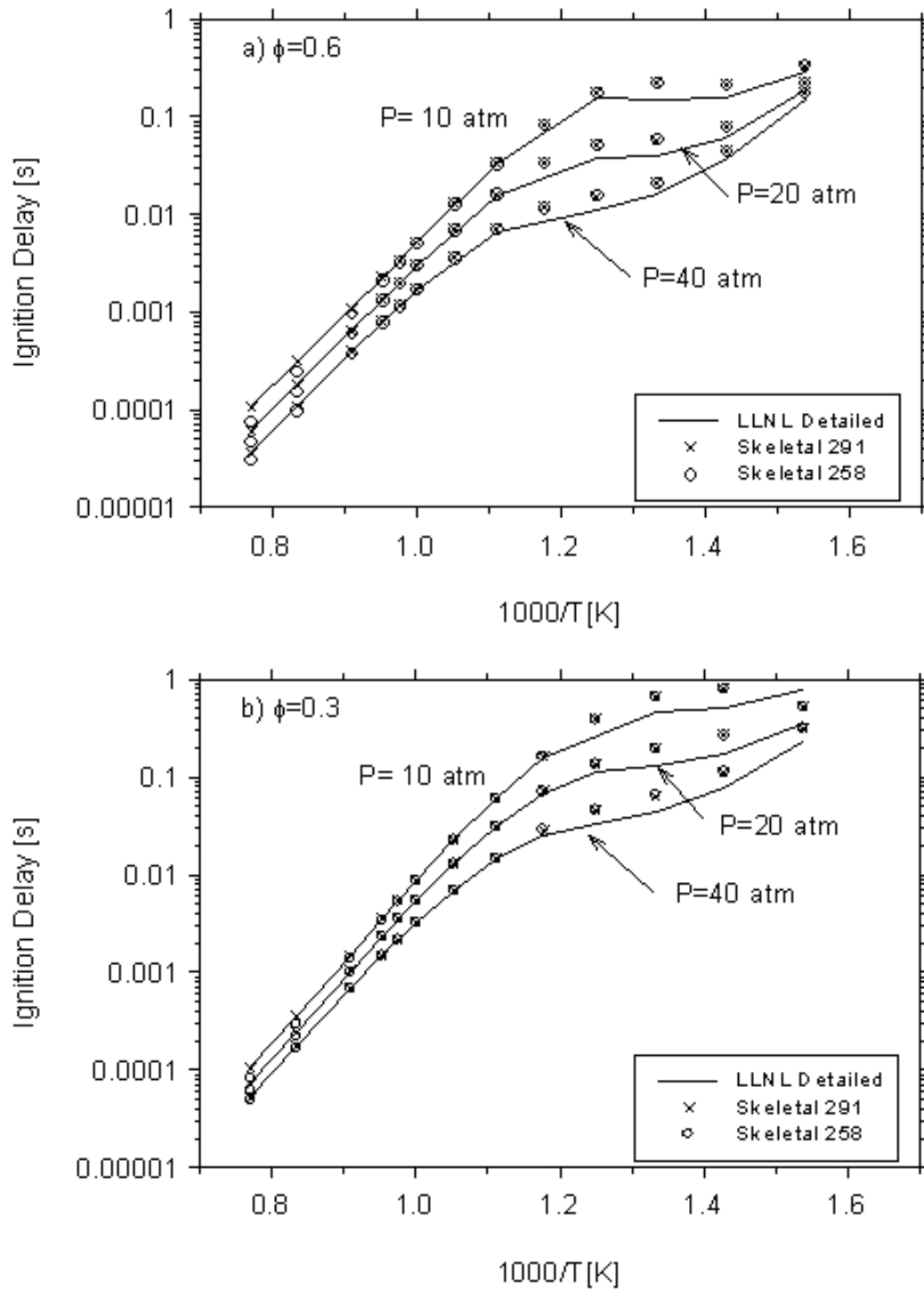


Figure 1. Comparison of predicted ignition delay times versus $1000/T$ at constant pressure using detailed LLNL mechanism and two skeletal mechanisms: a) $\phi=0.6$; b) $\phi=0.3$ under $P=10, 20$ and 40 atm. Both skeletal mechanisms yield results in excellent agreement with those from LLNL detailed mechanism for $T \geq 1,000$ K.

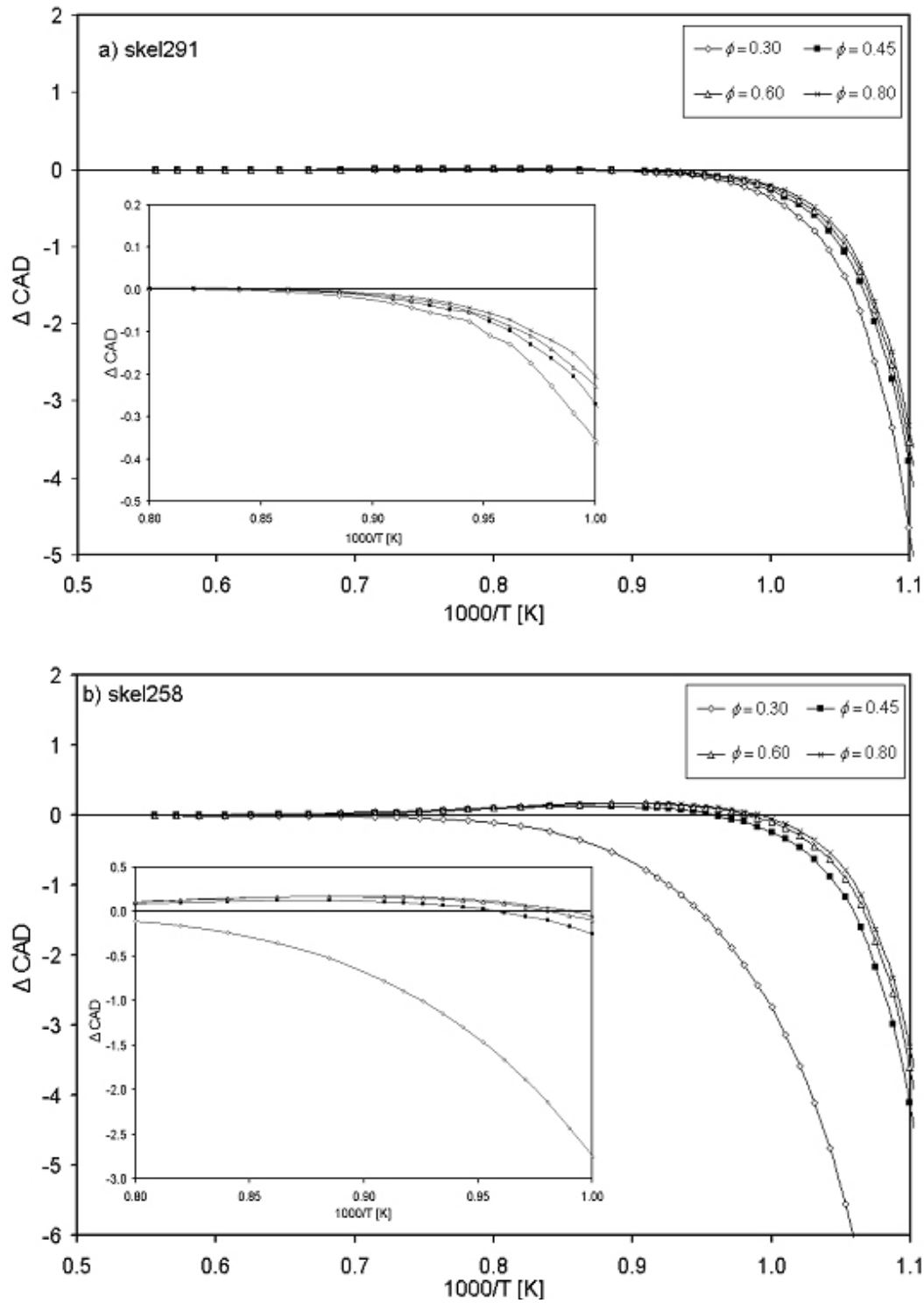


Figure 2. Comparison of crank angle degree difference (ΔCAD) at $P=40$ atm with $\phi=0.3, 0.45, 0.60,$ and 0.80 : a) ΔCAD between skeletal291 and LLNL detailed mechanism; b) ΔCAD between skeletal258 and LLNL detailed mechanism. Skeletal291 shows better agreement with those from LLNL detailed mechanism than skeletal258 especially for low equivalence ratios.

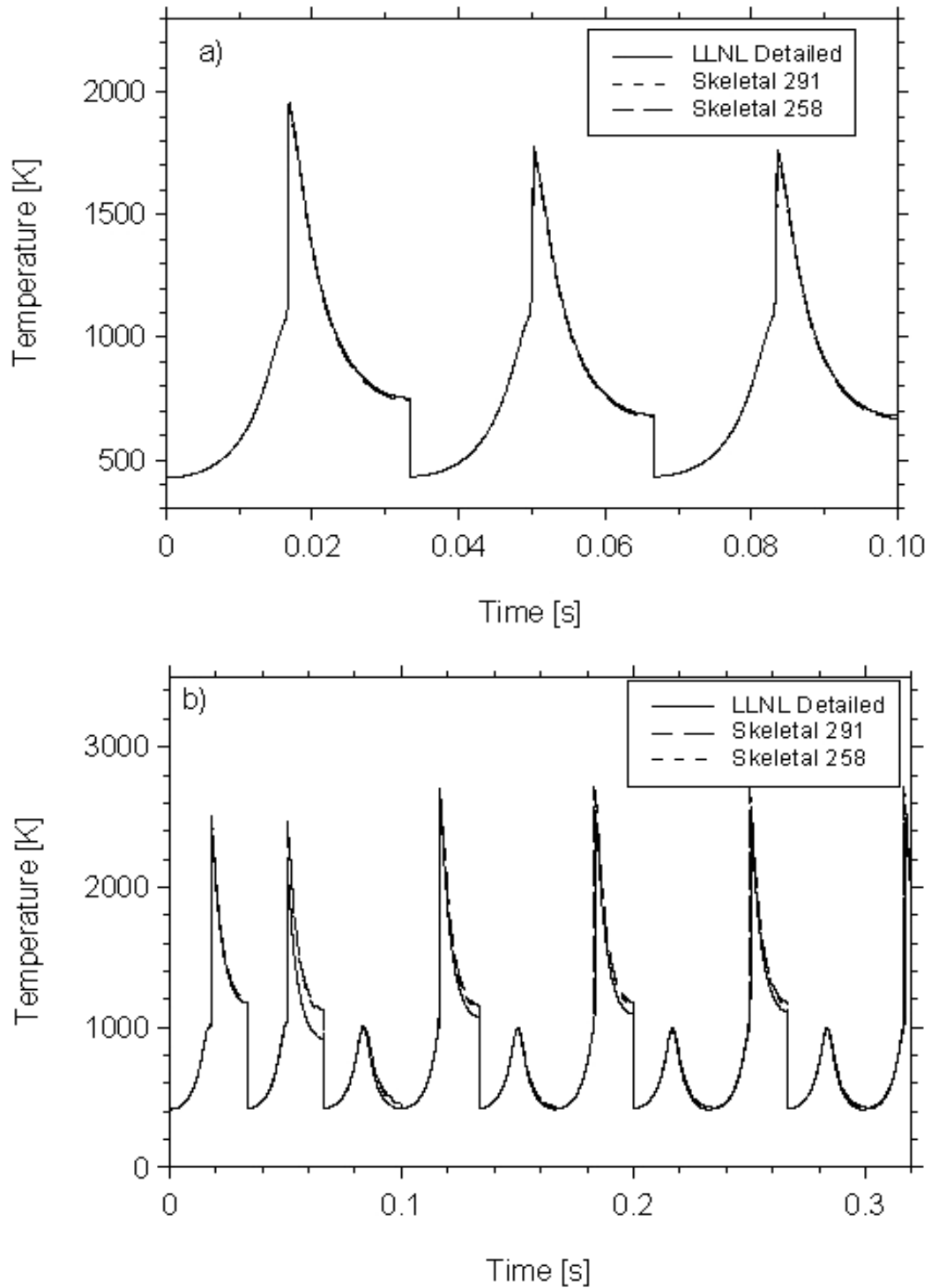


Figure 3. Comparison of transient HCCI simulations with temperature at BDC = 150°C, P = 1 atm: a) first cycle $\phi=0.3$ EGR=0, second and third cycles $\phi=0.3$ EGR=0.3 b) first cycle $\phi=0.6$, EGR=0, cycles after first $\phi=1$, EGR=0.6. Both skeletal mechanisms yield results in good agreement with those from LLNL detailed mechanism.

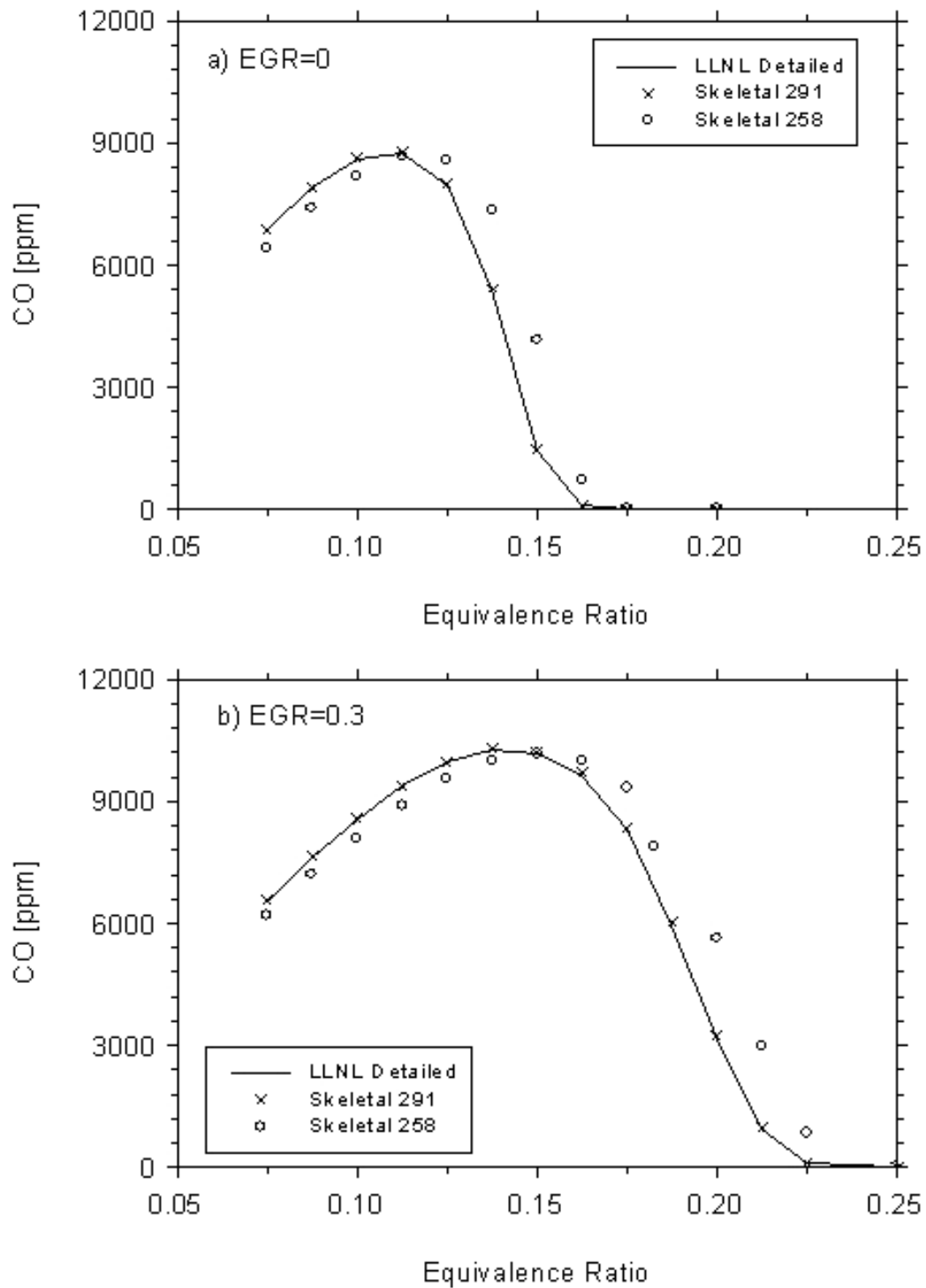


Figure 4. Comparison of predicted CO mole fractions [ppm] versus equivalence ratio with different mechanisms for engine parameters in Table 1: a) without EGR; b) with EGR=0.3. Skeletal291 gives results in much better agreement with LLNL detailed mechanism than skeletal258.

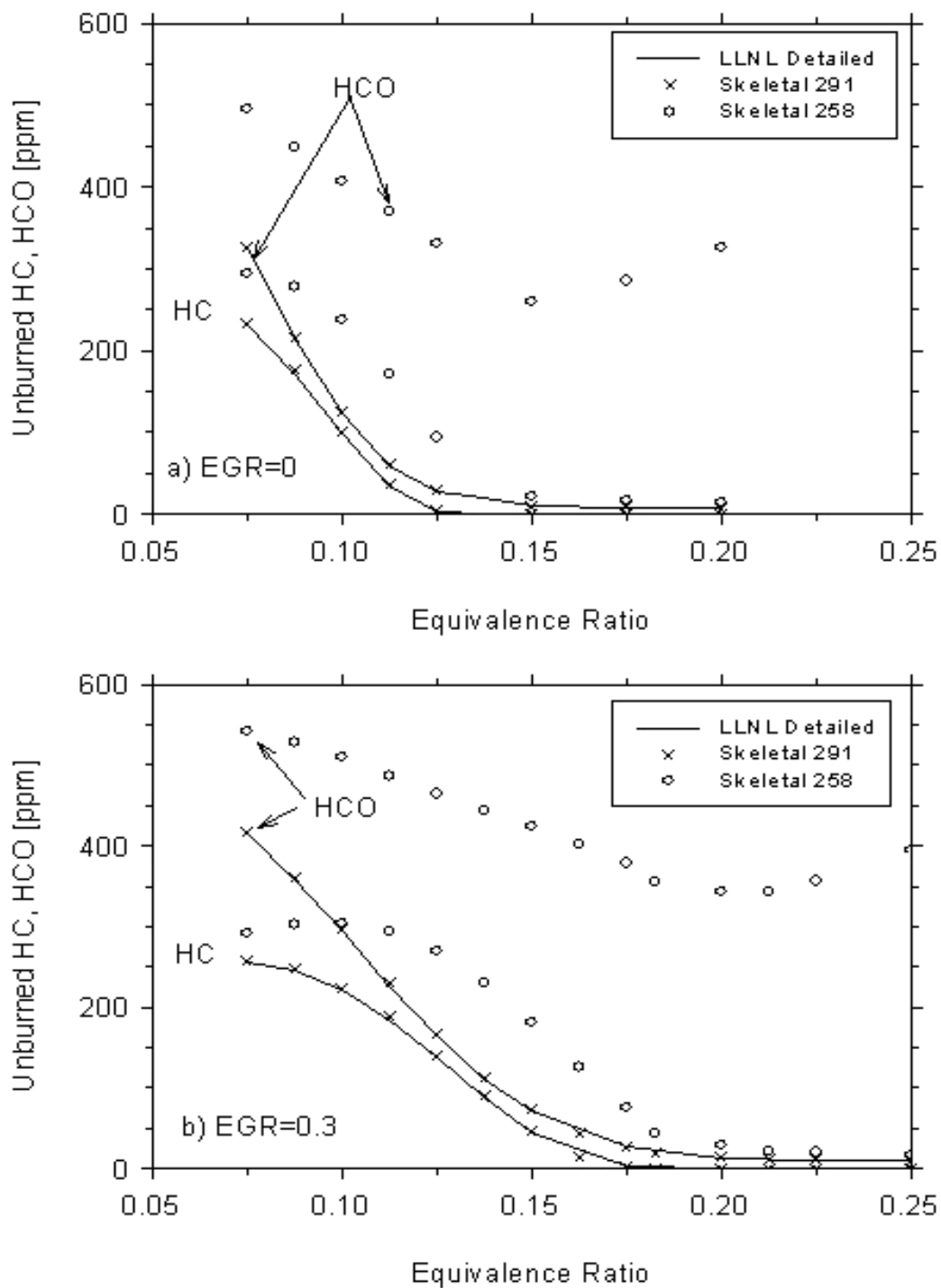


Figure 5. Comparison of unburned HC and HCO (total oxygenated unburned hydrocarbons) versus equivalence ratio for Table 1 engine conditions: a) without EGR (top); b) with EGR=0.3. Skeletal291 gives results in excellent agreement with LLNL detailed mechanism. Results from Skeletal258 contain large errors.