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A reduced mechanism for predicting the ignition timing of a fuel blend of natural-gas and n-heptane in HCCI engine

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ABSTRACT

One of the main challenges associated with homogeneous charge compression ignition (HCCI) combustion engine application is the lack of direct control on ignition timing. One of the solutions to this problem is mixing two fuels with various properties at a variety of ratios on a cycle-by-cycle basis. In the current study, a reduced mechanism for a fuel blend of natural-gas and n-heptane is proposed. The approach is validated for the prediction of ignition timing in the HCCI combustion engine. A single-zone combustion model is used to simulate the HCCI engine. A two-stage reduction process is used to produce two reduced mechanisms of existing semi-detailed GRI-Mech. 3.0 mechanism that contains 53 species and 325 reactions and Golovichev's mechanism consisting of 57 species and 290 reactions for natural gas and n-heptane fuels, respectively. Firstly, the unimportant species and related reactions are identified by employing the directed relation graph with error propagation (DRGEP) reduction method and then, to extend reduction, the principal component analysis (PCA) method is utilized. To evaluate the validity of the reduced mechanism, representative engine combustion parameters such as peak pressure, maximum heat release, and CA50 are used. The reduced mechanism of GRI-Mech. 3.0 mechanism, containing 19 species and 39 reactions, and the reduced mechanism of Golovichev's mechanism, consisting of 40 species and 95 reactions, provide good prediction for the mentioned parameters in comparison with those of detailed mechanisms. The combination of the generated reduced mechanisms is used to develop a reaction mechanism for a fuel blend of natural-gas/n-heptane. Then, the genetic algorithm is used for optimization of reaction rate constants in the newly generated mechanism. Simulation results agree well with the experimental results under various operating conditions, while maintaining small errors (less than 2° CA) for the mentioned engine combustion parameter. The proposed mechanism, which includes 41 species and 109 reactions, and two generated reduced mechanisms (for natural gas and n-heptane) are available as Supporting Information for this article.

1. Introduction

Two motivations drive the development of alternative energy technologies for the IC engines: the reduction of transportation emissions and fuel consumption. The homogeneous charge compression ignition combustion engine is a potential candidate for higher thermal efficiency and lower emissions [1–3]. Other major advantages of HCCI engines include their ability to burn with multiple fuels with various physical and chemical properties [4] and their ability to employ a combination of different fuels as blends, such as n-butanol/n-heptane, natural gas/n-heptane, and ethanol/gasoline [5–7]. However, these engines have two significant limitations: first, controlling the ignition timing, and second, their limited operational range [8–10].

Fuel blending is one of the approaches utilized for controlling HCCI combustion timing. Mixture ignitability can be adjusted on a cycle-by-cycle basis by mixing fuels with various ignition properties and altering the blend ratio. Examples of such controlling method are given as follows:

Christensen et al. [11] used a variable compression ratio technique accompanied with different inlet temperature, various blends of n-heptane/iso-octane, and regular gasoline/diesel blends to adjust the start of combustion angle to the set points for a single-cylinder HCCI engine. In order to obtain auto-ignition at around TDC for a blended fuel of n-heptane/iso-octane with higher octane number, the engine should adjust to work with greater compression ratio. This work demonstrated that HCCI engine with variable compression ratio can run almost on any liquid fuel.

By altering the proportion of ethanol and n-heptane in the mixture, Olsson et al. [12] controlled the combustion timing for a given load and thereby extended the operating range for a turbocharged

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Nomenclature

Greek

ϕ	Total equivalence ratio

Abbreviations

IVC	inlet valve closing
rpm	revolutions per minute
TDC	Top dead center
EGR	exhaust gas recirculation
SOC	start of combustion
NG	Natural gas
SZCM	single zone combustion model
IC	internal combustion
ODE	ordinary deferential equation
$\tilde{F}$	log-normalized rate sensitivity matrix
GA	genetic algorithm
MHR	maximum heat release
CO_2	carbon dioxide
HC	hydro-carbon
BMEP	brake mean effective pressure
IMEP	indicated mean effective pressure

HCCI engine. Results indicate that at low loads, the ratio of n-heptane in the mixture was increased to advance the combustion timing, while this manner changes as the load increases.

Hosseini et al. [13] showed that adding Reformer gas (RG) to the n-heptane fuel causes reduction of heat release in the first stage of combustion in the well-known two-stage combustion of n-heptane fuel. It also shifts the second stage of combustion to a more optimized crank angle position, which increases the indicated power and fuel conversion efficiency.

Nathan et al. [14] studied the possibility of using the HCCI technology to exploit biogas effectively in IC engines. Biogas has a high self-ignition temperature and used as the main source of energy. Therefore, diesel fuel, with a low self-ignition temperature was blended in for improved ignition and to control the start of combustion. The work demonstrated that the biogas–diesel HCCI mode can work at efficiencies close to that of diesel operation while attaining extremely low levels of NO and smoke in a BMEP range of 2.5–4 bar. However, the amounts of HC emissions of biogas–diesel HCCI mode are significantly higher in comparison with the normal diesel mode.

Accurate fuel oxidation chemistry models of such a blended fuels offer great potential for HCCI engine design and optimization. However, there are fewer publications on studying of blended fuel combustion.

Brakora et al. [15] developed a reduced mechanism for combustion characteristics prediction of diesel/biodiesel blended fuel in a HCCI engine. The reduced mechanism of methyl butanoate, which was generated by applying reduction methods such as flux analysis, ignition sensitivity analysis, and optimization of reaction rate constants, was combined with the reduced mechanism for n-heptane oxidation. Reaction constants of specific reactions in the combined mechanism were then adjusted for the single zone combustion model to improve the performance of the mechanism for prediction of the ignition delay time.

Dagaut and Togbé [16] developed a detailed chemical mechanism of butanol/gasoline mixture by a combination of kinetic schemes for the oxidation of the pure components of the butanol/gasoline surrogate. In another work, Dagaut and Togbé [17] performed a kinetic modeling of ethanol/n-heptane mixtures oxidation by merging the kinetic mechanisms of n-heptane fuel and an ethanol oxidation sub-scheme. They showed that utilizing the resulting comprehensive chemical kinetic mechanisms in perfectly stirred reactor (PSR) systems have good accuracy in predicting the mole fractions of the fuel components and of the main products.

Most recently, Aggarwal et al. [18] studied the ignition behavior of heptane/methane fuel blends at conditions relevant to diesel/HCCI engines in a closed homogenous reactor. They showed that the termed Chalmers mechanism [19], consisting of 42 species and 168 reactions, agreed with the shock tube ignition data for the mixtures of pure n-heptane with air and also for the mixture of pure methane with air. As a result, this mechanism has been selected to investigate the ignition behavior of n-heptane/methane fuel blends. It has been shown that the addition of n-heptane decreases the ignition delay for methane-air mixtures in both low and high temperature conditions. However, the authors have not provided any validation with respect to experimental data related to a natural-gas/n-heptane blend fueled HCCI engine.

The objective of the present study is to produce a valid reduced mechanism of a fuel blend of n-heptane/natural-gas for prediction of start of combustion (SOC) in the HCCI engine. Natural gas has been selected since it is mostly used in diesel engines as dual fuel form, both to improve the performance [20,21] and because it is suitable for preparing homogenous mixture. According to Zheng and Caton [22,23] the GRI-mech. 3.0 [24] was adopted to simulate a NG fueled HCCI engine. In agreement with Hosseini et al. [13], the selected mechanism for n-heptane fuel is the Golovichev's mechanism [19]. Chemical kinetic combustion models are built in a hierarchical manner. Therefore, a detailed model for n-heptane is expected to have a validated sub-mechanism for methane. However, the simulation results shows that this mechanism failed to properly predict experimental ignition timing of a fuel blend of n-heptane/natural-gas at different operating conditions. The Curran mechanism which is even more comprehensive, with 560 species and 2539 reactions, generates important discrepancies with respect to experimental results for the blend fuel. A similar behavior was observed by Kongsereeparp [25] when they tried to use the n-heptane mechanism for predicting the ignition timing of a fuel blend of n-heptane and reformer gas (75%H2–25%CO) for HCCI condition, although the selected mechanism was in good agreement with the shock tube ignition data.

In the present work, a two-stage reduction processes is utilized to generate two reduced mechanisms of natural gas fuel and n-heptane fuel. In the second step, the final reduced mechanism for a natural-gas/n-heptane blend is proposed by combination of these two mechanisms and optimization of the reaction rate parameters utilizing genetic algorithm. Ignition timing data for the n-heptane/natural-gas mixture are used to validate several reaction mechanisms for these mixtures.

2. Methods of mechanism reduction

The two main procedures used for reducing the detailed chemical kinetic mechanisms are known as time scale analysis and skeletal reduction. The objective in both approaches is to identify and remove the redundant species and reactions with the intention of reducing the computation time in simulations. The results obtained are approximately the same as those obtained by the corresponding detailed mechanism. Examples of time scale analysis include computational singular perturbation (CSP) [26] and intrinsic low-dimensional manifolds (ILDM) [27] methods. On the other

hand, skeletal reduction contains sensitivity analysis [28], principal component analysis (PCA) [29], optimization-based techniques [30,31], element flux-based method [32], directed relation graph (DRG) [33] and also directed relation graph with error propagation (DRGEP) [34,35]. Furthermore, dynamic methods like the work of He et al. [36] and Liang et al. [37,38] update the reduced mechanism dynamically at each time step based on the local conditions, and thereby develop locally accurate mechanisms. However, as pointed out in Ref. [36], the discontinuity in the conversion rate of species, when mechanisms switch from one type to another during the simulation in the fly scheme, may cause composition oscillations of species. This may lead to failure in solving the governing ODEs.

Multi-stage reduction schemes, which are the integration of two or more reduction methods, can be used to increase the extent of reduction such as DRGASA [39], DRGEP-PCA [40], and DRGEP-CSP-DRGEP [41]. The present work utilizes the integration of DRGEP and PCA methods.

It is observed that when the DRGEP method is used to vary the concentration of species or completely eliminate them, the error term will be restrained during the process of reaching the target species [34,35]. Target species are the group of species determined to be of interest to the investigator. Species that are not closely associated with the target are considered less important.

As mentioned by Vajda et al. [42] the kinetic information inherent in the elements of the log-normalized rate sensitivity matrix $\tilde{F}$ can be extracted by principal component analysis. In such calculations, $\tilde{F}$ is taken as the ratio of the rate of formation or consumption of species i in reaction j and the net rate of the concentration change of species i. Kinetic information comes by performing eigenvalue–eigenvector of the matrix $\tilde{F}^T\tilde{F}$. The important reactions can be defined as the significant eigenvector elements of reactions, which are characterized by large eigenvalues. By providing the user-specified tolerances for these parameters, unnecessary reactions can be identified.

The SZCM and reduction procedure used in this work for generating the reduced mechanism is almost similar to the previous paper of the authors [41] except that PCA is being used instead of CSP. This paper does not attempt to investigate the advantages/disadvantages of these two methods since authors plan to examine this issue in a future study.

3. Experimental set-up and engine specifications

All experiments were carried out in the engine research facility of University of Alberta by using a Waukesha CFR single cylinder engine coupled to a DC dynamometer. Engine specifications were presented in Table 1. The experimental set up is shown in Fig. 1.

The engine was run with an open throttle at a constant speed of 800 RPM. A 2.4 kW heater with PID temperature controller was used to pre-heat the intake air when required. Two types of fuel injectors, one for the n-heptane and the other for natural gas (NG), were located upstream of the intake valve to facilitate proper mixing. The control module of an AFS Sparrow-II engine was utilized to regulate the injection rate of each fuel separately or as a blend. External uncooled exhaust gas recirculation (EGR) line was connected to the intake after the heater and before the fuel injectors and was controlled by a manual butterfly valve. Intake gas was analyzed for the EGR fraction determination. A Vetronix PXA-1100 portable gas analyzer which is capable of measuring different gases was used in the intake system and EGR connection to the intake plenum to determine the CO_2 concentration in the intake mixture.

EGR was calculated using volume concentration measurement of CO_2 upstream and downstream of the engine. EGR was calculated as:

$$\mathrm{EGR} = 100 \times \frac{\mathrm{CO}_{2;up}}{\mathrm{CO}_{2;down}} \tag{1}$$

To measure pressure signal in the combustion chamber on a resolution of 0.1 CAD, a Kistler 6043A pressure transducer was used. Experiments were monitored with the help of a personal computer with Labview software installed on it. Three NI PCI-MIO-16E1 data acquisition cards with high sampling rates were used with the computational system.

The intake pressure at the IVC time was taken as the reference point for the pressure trace signal. The high frequency noise was filtered digitally from the pressure trace signal. The pressure traces measured over 100 successive cycles were averaged to avoid cyclic variations before they were used in calculations. The different cases studied with this experimental set up are shown in Tables 2–4.

Table 1
Engine specifications.

Parameter	Specification
Engine model	Waukesha CFR
Engine type	Water cooled, single cylinder
Combustion chamber	Disk cylinder head, flat-top piston
Throttle	Fully open
Bore	82.6 mm
Stroke	114.3 mm
Displacement	612 cc
IVC	146 CAD BTDC
EVO	140 CAD ATDC

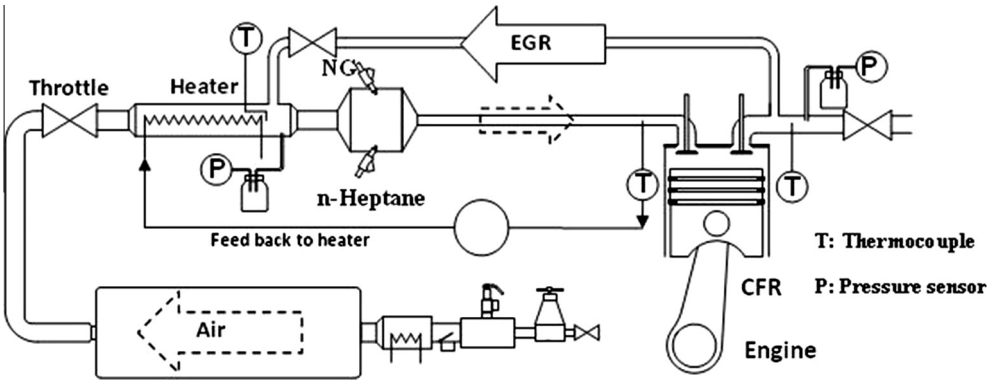

Fig. 1. Schematic of the engine lab hardware.

The parameters used in the present work were sorted into three groups, namely, primary, secondary and cyclic related. The error analysis was conducted differently for each group.

In the primary group, air and fuel flow rates, all measured temperatures, concentrations of exhaust gas species and intake and exhaust pressures were taken into account. Internal error analysis was performed for these parameters. In a steady-state engine operation the measurements were repeated 20 times for each parameter in this category.

The secondary parameters are defined to be those variables which were calculated from the primary parameters. Examples are indicated power and thermal efficiency. External error analysis was utilized for this group of parameters.

A reasonable number of the parameters, such as SOC, Pmax etc., presented in the present study were averaged over 100 consecutive cycles. Cyclic variation in the HCCI combustion is less than that of the conventional combustion, but as far as the absolute estimated error from internal and external errors is concerned the deviation is significant. Averaging over 100 repeated cycles, the cyclic error related to each parameter was predicted. The uncertainty for engine parameters and combustion parameters are displayed in Table 5.

Table 5
Experimental uncertainty.

Uncertainty	Parameter
<1%	Temperature
<1%	Crank angle position
<2%	Engine speed
<1%	Fuel Flow rate
<1%	Air flow rate
<1%	EGR
<2%	IMEP
<2°	SOC
<2%	Pmax
<2%	MHR
<2°	CA50

4. Genetic algorithm

As it will be discussed later, an optimization of reaction constants is required for the blended fuel in the proposed mechanism. This mechanism is the combination of generated reduced mechanism of natural gas and reduced mechanism of n-heptane. The present study uses a Fortran programming code for GA algorithm implementation. The algorithm fetches the inputs from a file preparing a population randomly in the range of +30% and −20% of the original values. By utilizing the original population, it then recalls SZCM and makes a primary evaluation of fitness function.

The GA is put into practice by binary coding, tournament selection and functions computation procedures with weighting factors which are selected depending on the functions importance [43]. The definition of the Fitness function is:

$$\text{Fitness function} = \sum_{i=1}^{n}\left[W_i \times \left|\frac{X_{i,code} - X_{i,test}}{X_{i,test}}\right|\right] \quad (2)$$

The term $\left|\frac{X_{i,code}-X_{i,test}}{X_{i,test}}\right|$ is the ith target function in which, $X_{i,code}$ is an estimation of the ith variable by using engine simulation model and $X_{i,test}$ represents the ith variable obtained experimentally. In the case that more than one target functions are selected for optimization, W_i stands for weight coefficients of such parameters. However, in this work W_i is equal to 1. The SOC was selected as the target parameter in this work. The main purpose of this study is to produce a reduced chemical kinetic mechanism that provides the most accurate simulation of the ignition timing for natural gas and n-heptane blend fuel HCCI combustion. Timing of the SOC is an important feature of the HCCI engine. When the ignition occurs too soon in the engine cycle, excessive rates of pressure-rise are produced, which may result in high combustion noise or engine damage. On the other hand, the combustion may be quenched because of too late ignition timing resulting in large amount of HC emission and no work output.

The following advantages are also achieved by selecting the ignition timing as the main feature of HCCI combustion characteristic:

(1) It can be computed with single-zone modeling. (2) It is a single output obtained from all the chemical reactions taken into account. (3) It can be correctly determined both experimentally and by simulations.

It would be more rational to use single-zone modeling in GA optimization, since it is not only very accurate but also it takes a lot less computer runs than a multi-zone model. Besides, it is not

Table 2
Operating conditions for considered cases of natural gas.

Case	1	2	3	4	5
Equivalence ratio (Φ)	0.69	0.53	0.45	0.45	0.42
NG mass rate (mg/s)	107	95.49	92.43	92.43	85.59
Air mass rate (g/s)	2.51	2.94	3.35	3.30	3.33
T_{IVC} (K)	413	413	413	413	413
P_{IVC} (bar)	1.57	1.56	1. 56	1. 56	1.56
% EGR	41.1	30.5	20.7	22.2	22.2
Compression ratio	17.25	17.25	17.25	17.25	17.25

Table 3
Operating conditions for considered cases of n-heptane.

Case	1	2	3	4	5
Equivalence ratio (Φ)	0.68	0.41	0.43	0.38	0.26
n-heptane mass rate (mg/s)	103.43	75.52	89.69	92.40	79.23
Air mass rate (g/s)	2.28	2.77	3.11	3.66	4.57
T_{IVC} (K)	333	333	413	413	413
P_{IVC} (bar)	1.54	1.55	1.56	1.54	1.57
% EGR	51.01	40.69	31.66	19.79	0.0
Compression ratio	11.5	11.5	11.5	11.5	11.5

Table 4
Operating conditions for considered cases of natural gas/n-heptane blend fuel.

Case	1	2	3	4	5	6	7	8	9
Equivalence ratio (Φ)	0.44	0.43	0.61	0.32	0.48	0.91	0.76	0.63	0.86
NG mass rate (mg/s)	21.70	28.68	32.49	18.86	24.18	45.97	42.25	40.33	29.79
n-heptane mass rate (mg/s)	37.74	35.57	40.37	33.63	39.99	41.06	39.36	36.94	49.26
Air mass rate (g/s)	2.08	2.33	1.87	2.52	2.07	1.50	1.68	1.90	1.43
T_{IVC} (K)	393	393	393	393	393	393	393	393	393
P_{IVC} (bar)	1.01	1.00	1.00	1.00	1.00	1.01	1.01	1.01	1.01
% EGR	19.19	8.14	23.79	0.0	19.02	34.56	28.03	21.81	37.38
Compression ratio	13.8	13.8	13.8	13.8	13.8	13.8	13.8	13.8	13.8

possible to use the multi-zone model with the presently existing computer technology.

The optimized mechanism is obtained as the defined fitness function approaches to minimum. Other GA parameters used include the population number, crossover probability, mutation probability and maximum generation. In this study, these parameters are set to 50, 0.5, 0.02 and 1000, respectively. A total of 9 cases displayed in Table 4 are examined in this study. Four different engine operating conditions (Cases 1–4) have been selected for the optimization of reaction rate parameters by applying the GA. Five different engine operating conditions (cases 5–9) have been chosen to investigate the validity of the combined chemical kinetics mechanism presented here.

5. Results and discussion

5.1. Mechanism reduction

The GRI-Mech. 3.0 [24] and an updated version of Golovichev's [19] kinetic reaction mechanisms, consisting of 53 species and 325 reactions and 57 species and 290 reactions respectively, are chosen for the oxidation of natural-gas (NG) and n-heptane fuels.

Fig. 2 demonstrates the validity of the single zone combustion model utilized in this work in capturing cylinder pressure experiment data through the compression and SOC process for n-heptane fueled HCCI engine and natural gas fueled HCCI engine at some selected operating conditions. It is clear that the applied single zone combustion model is in good agreement with experimental data and the error in prediction of SOC is less than 2° CA. Since the Golovichev's mechanism already includes the sub-mechanism for methane, the validity of this mechanism is questioned for the blend fuel of n-heptane and natural gas. Fig. 3 shows a comparison of predicted in-cylinder pressure traces during the compression stroke resulting from the single-zone combustion model for n-heptane and natural gas blends combustion in HCCI engine with the corresponding experimental data for some selected operating conditions. It is clear that by using this mechanism, the predicted in-cylinder pressure cannot follow the corresponding experimental data as well as SOC. Fig. 3 also points out that the Curran mechanism, with 560 species and 2539 reactions, shows remarkable discrepancies in the above mentioned parameter with respect to the experimental results for the blend fuel.

As a result, in this work, two reduced mechanism of n-heptane and natural gas fuels (which represent the main features of the corresponding detailed mechanism) are generated, and a combination of these two mechanisms is tested to represent the combustion of a fuel blend of n-heptane and natural gas. In the first stage, two reduced mechanisms of n-heptane and natural gas are developed. Five different engine operating conditions are selected for combustion analysis for each of natural gas and n-heptane fuels listed in Tables 2 and 3, respectively. The reduction process, which is used in the present work, is based on a two-stage reduction method which utilizes DRGEP and PCA methods, successively. More specifically, DRGEP identifies and eliminates unimportant species and reactions. Then, in the second stage, the PCA method is applied to improve the process by eliminating redundant reactions. For DRGEP reduction, like Liang et al. [37] and Shi et al. [40], fuel, HO_2, and CO are selected as the target species at each sampling point. The reasons for this selection are discussed in detail in Ref. [37]. Species that are reachable from the target species are identified at each sampling point and the collection of all of these species sets constitutes the final important species set for a specific operating condition. The rest of the species are considered unimportant

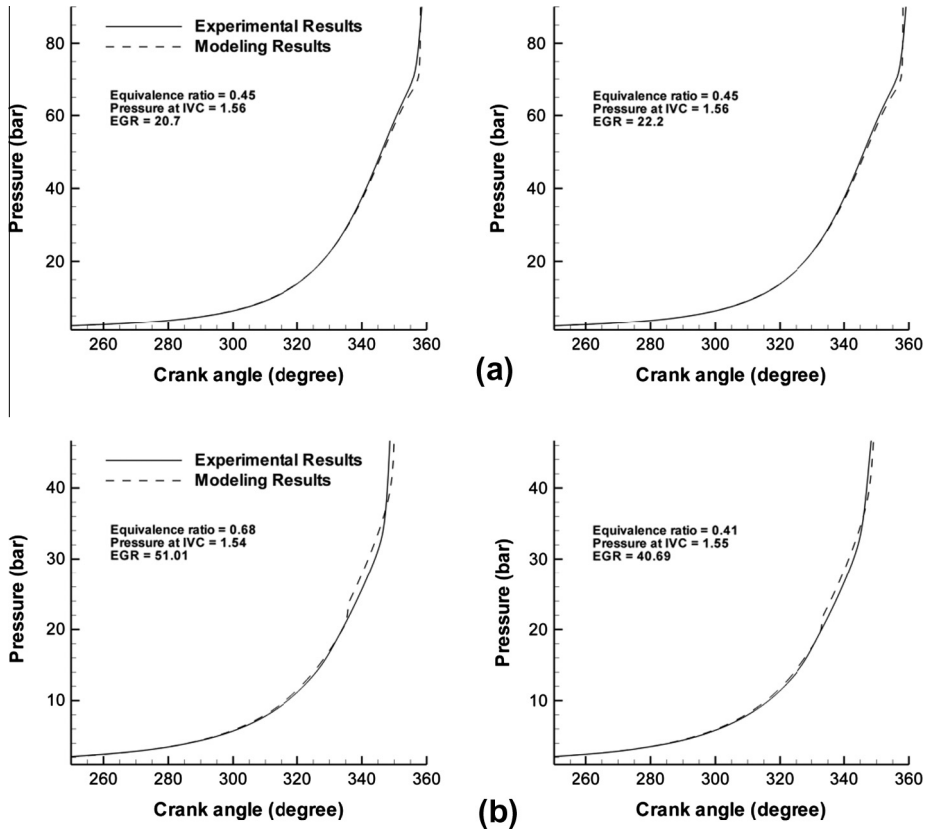

Fig. 2. Comparison of predicted in-cylinder pressure traces during the compression stroke resulting from the single-zone combustion model with the corresponding experimental data (a) pure natural gas fuel and (b) pure n-heptane fuel.

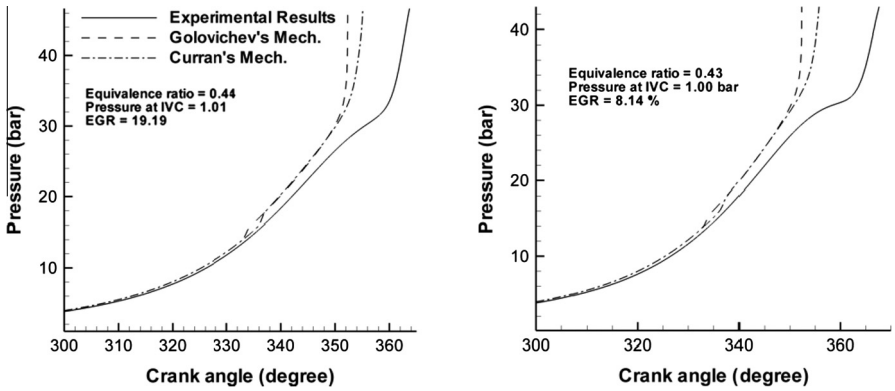

Fig. 3. Comparison of predicted in-cylinder pressure traces during the compression stroke resulting from the single-zone combustion model utilizing the Golovichev's mechanism and the Curran's mechanism with the corresponding experimental data.

species, and reactions that include any of these species are eliminated from the final mechanism.

An initial tolerance value to start the DRGEP reduction process is 10^{-5} and the tolerance value for PCA reduction process that follows the DRGEP process is 10^{-3}. The selected representative parameters for investigating the validity of the reduced mechanism in this work are the predicted CA50, peak pressure, and maximum heat release. At each generation, these parameters are calculated using the HCCI engine simulation code and compared to the results obtained by using the detailed mechanism for the considered cases. The generated reduced mechanism is considered as valid one if the errors in predicted CA50, peak pressure, and maximum heat release using reduced mechanism do not exceed 1° CA, 1%, and 1% respectively, with those of the detailed mechanism. Therefore, for each operating condition mentioned in Tables 2 and 3, the developed reduced mechanisms are in different final sizes as can be seen in Table 6.

Figs. 4 and 5 show the mechanism size and the interfered errors in calculation of CA50, peak pressure, and maximum heat release because of elimination of insignificant species and reactions and algorithm error tolerances at each generation during DRGEP and PCA reduction processes, which are distinguished with a vertical line. The considered cases in these figures are test condition 5 and test condition 2 for natural gas and n-heptane fuels, respectively. For the first reduction stage, which utilizes DRGEP reduction method, insignificant species and their corresponding reactions are identified and eliminated effectively from the mechanism. Following the first stage, PCA reduction is applied to the mechanism to remove further reactions in the second stage. By applying this two-stage reduction method, a detailed mechanism used for simulating the combustion of natural gas comprising 53 species and 325 reactions, is reduced to a smaller mechanism containing 19 species and 39 reactions. Also, a detailed mechanism of n-heptane including 57 species and 290 reactions is cut down to a more concise mechanism consisting of 40 species and 95 reactions. The reduced mechanisms (generated for case 5 of natural gas-fueled HCCI engine and case 2 of n-heptane-fueled HCCI engine) are provided as additional information for the present work.

By utilizing the specified reduction processes for each of the operating conditions, the corresponding reduced mechanism is generated. However, it is required to have a single reduced mechanism. Normally, this final reduced mechanism can be developed by the combination of all the generated mechanisms for each case. Another alternative solution for this goal, as mentioned in Ref. [40], and also can be seen in Figs. 6 and 7, is to evaluate the performance of each of the reduced mechanisms at different operating conditions. It can be seen that most of the generated mechanisms can be used for all other cases while predicting the representative parameters in the error tolerance limit. For example, the generated reduced mechanism for case 1 for natural gas fuel is used to simulate the combustion phase of the natural gas-fueled HCCI engine in different considered cases in Table 2. The calculated error in predicting peak pressure, maximum heat release, and CA50 between reduced and detailed mechanisms are fewer than the user specified error tolerance values. However, the developed reduced mechanism of case 4 for n-heptane is unable to accurately predict some of these parameters for operating conditions of case 1, case 2, and case 3. Therefore it is not considered as a valid one.

To verify the ability of the reduced mechanism in predicting the pressure trace and heat release rate histories, a comparison of these parameters between reduced and detailed mechanisms are depicted in Fig. 8 for different operating conditions of both natural gas-fueled and n-heptane-fueled HCCI engine. As indicated in this Figure, simulation using reduced mechanism is in good agreement with the simulation utilizing detailed mechanism. Furthermore, the two-stage combustion behavior of n-heptane fuel is accurately captured by the reduced mechanism for all cases.

SOC calculated at 10% of total heat release was also evaluated for both reduced and detailed mechanism for comparison reasons in Fig. 9. Combustion in HCCI engines is governed by the chemical kinetics. It is observed that the SOC is predicted accurately for all considered cases.

Table 6
Comparison of natural gas and n-heptane skeletal mechanisms sizes generated at each operating conditions.

For natural gas fuel				For n-heptane fuel			
Case	Species	Reactions	Allowable error tolerances are exceed?	Case	Species	Reactions	Allowable error tolerances are exceed?
Case 1	19	37	No	Case 1	38	65	No
Case 2	19	36	No	Case 2	40	95	No
Case 3	19	41	No	Case 3	36	104	No
Case 4	19	35	No	Case 4	38	74	Yes
Case 5	19	39	No	Case 5	38	85	No

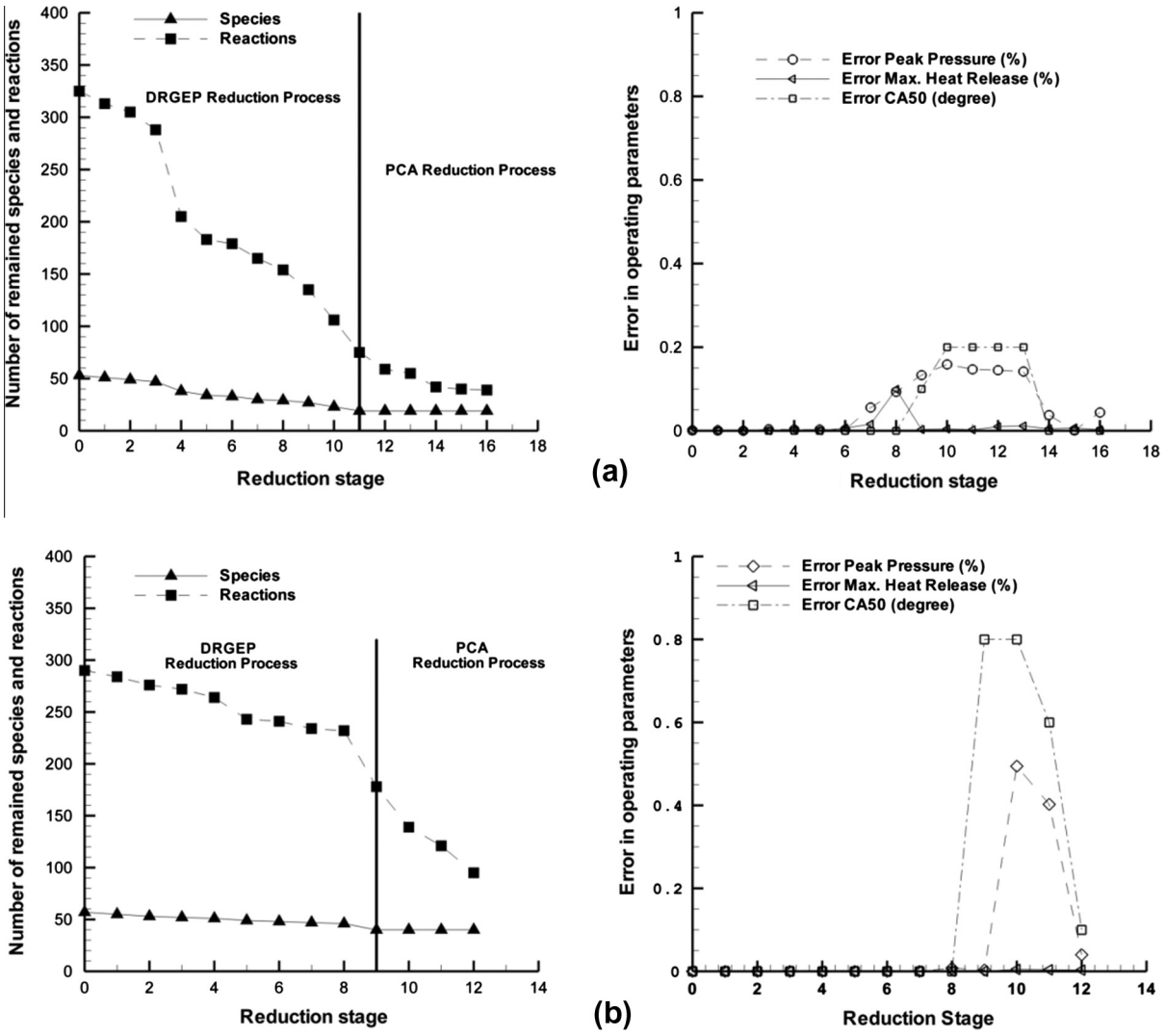

Fig. 4. Mechanism size and the corresponding error values at each reduction stage for (a) natural gas (case 5) and (b) n-heptane (case 2).

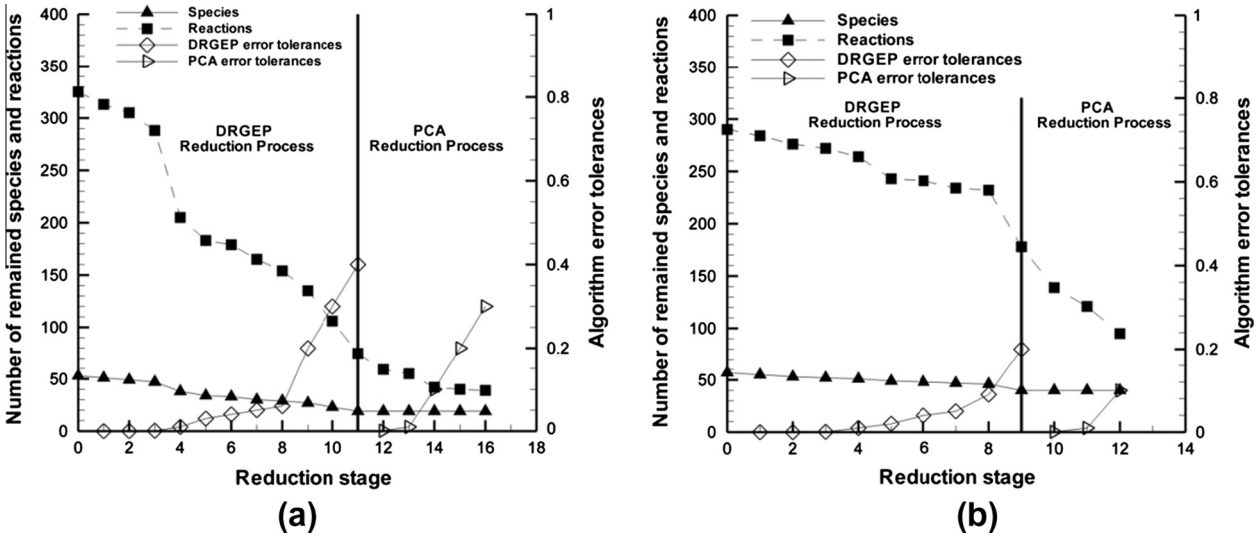

Fig. 5. Algorithm error tolerances. (a) for case 5 of the NG fueled HCCI engine and (b) for case 2 of the n-heptane fueled HCCI engine.

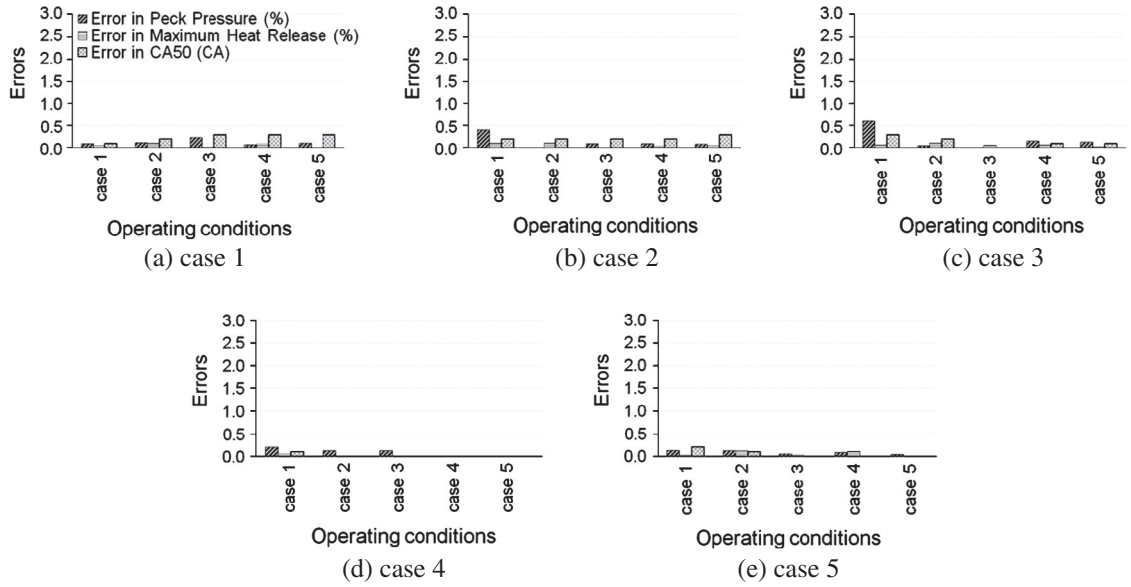

Fig. 6. Performance of each generated reduced mechanism for natural gas fuel at different operating conditions.

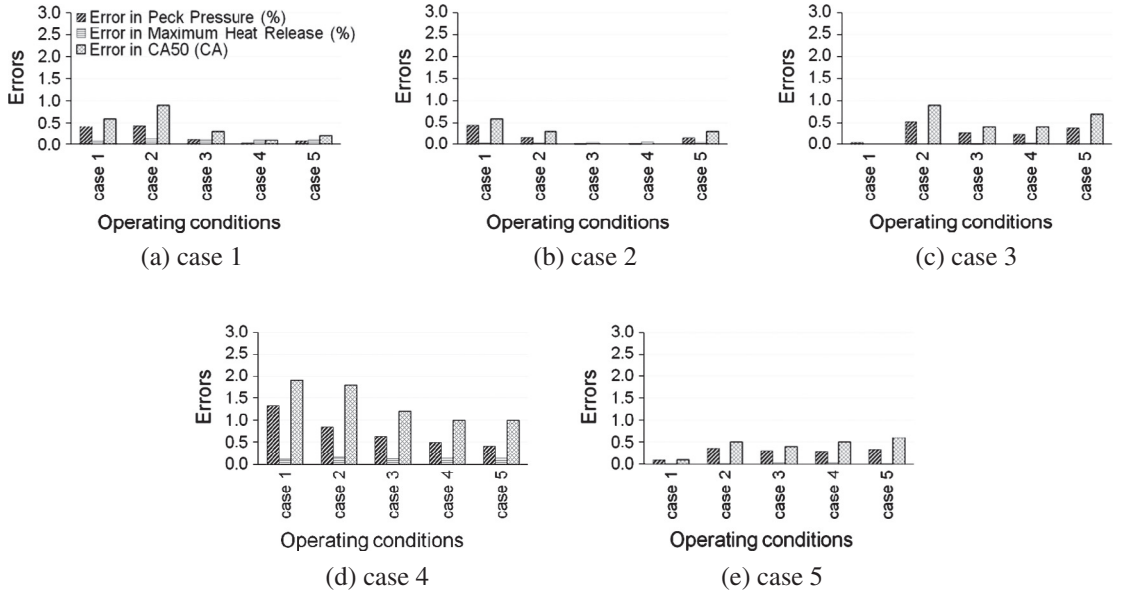

Fig. 7. Performance of each generated reduced mechanism for n-heptane fuel at different operating conditions.

Finally, a comparison of the simulation results for the mass fraction profiles of some major species such as fuels and CO using the reduced mechanisms and the detailed mechanisms are shown in Fig. 10. The results show that the calculated mass fraction of these species using the reduced mechanisms agrees very well with the simulation results utilizing the detailed mechanisms for both fuels.

5.2. Validation of combined chemical kinetics mechanism

As mentioned before, the purpose of this study is to generate a proper reduced chemical kinetic mechanism for oxidation of fuel blend of natural-gas and n-heptane using HCCI combustion model. To do this, two developed reduced mechanisms of n-heptane and natural gas are combined. The combined mechanism is based on natural gas mechanism in which species and corresponding reactions that are unique to the n-heptane mechanism are added to the natural gas mechanism. The resultant mechanism includes 41 species and 109 reactions. A set of different experimental conditions mentioned in Table 4 over wide ranges of equivalence ratios, intake pressures, and EGR rates are selected to evaluate the validity of this mechanism.

Fig. 11 displays the prediction of SOC for original combined mechanism. Significant disagreement is observed by utilizing original combustion parameters related to experimental results. Therefore, to improve the combustion prediction of the original mechanism, the optimization of the reaction rate constants is vital.

Genetic algorithm is used to optimize the constants of the reaction rate. A Fortran code was developed which firstly utilized the GA code to produce a population randomly and then, by recalling the SZCM, the start of the combustion was calculated for each of the considered operating conditions. Then an initial estimate of the fitness function was performed using this original population. This loop was repeated, if necessary, until the fitness function, the average error between the predicted and the measured ignition

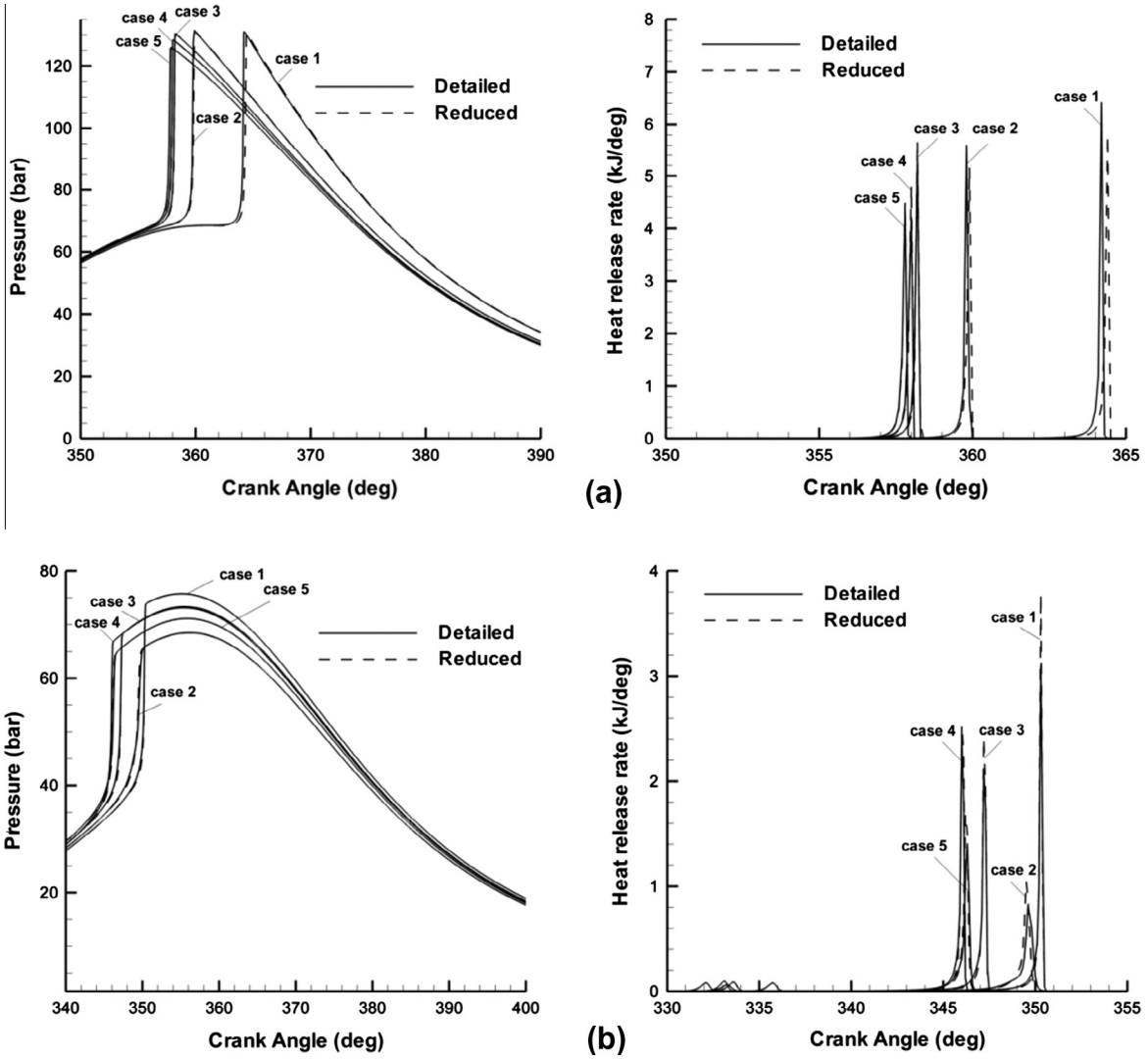

Fig. 8. Comparison of pressure traces and heat release rate histories (a) by applying the detailed GRI mechanism and its reduced mechanism generated for case 5 at different operating conditions and (b) by applying the detailed Golovichev's mechanism and its reduced mechanism generated for case 2 at different operating conditions.

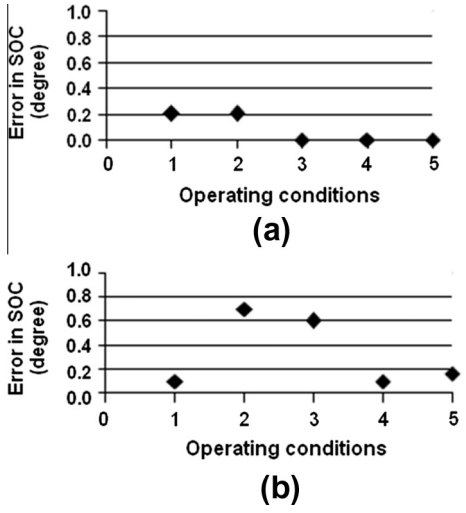

Fig. 9. Error in prediction of SOC of (a) natural gas and (b) n-heptane fuels in HCCI combustion engine for reduced mechanisms relative to the detailed ones at all considered cases.

timing over the entire operating conditions of interest, was less than a specified tolerance.

It should be noted that only the Arrhenius coefficients were changed in the optimization process. The other parameters such as the enhanced factors and the Troe parameters were unaltered from their values in the original mechanisms (i.e. the GRI-Mech 3.0 and Valeri mechanisms).

To optimize the constants of reaction rate, the four different operating conditions considered in Table 4 (cases 1–4) are applied to the GA. The optimized mechanism reduced the error in prediction of the ignition timing less than 2° CA for all of the operating conditions. To further examine the validity of the optimized mechanism, the deviation of simulation results of the reduced mechanism from experimental data for five different operating conditions presented in Table 4 (cases 5–9) are also investigated as shown in Fig. 11. This Figure demonstrates the differences between the predicted results for start of combustion by utilizing the optimized combined chemical kinetics mechanism and the experimental data. It can be seen that for all cases, this parameter is predicted accurately within the defined tolerance.

Fig. 12 shows a Comparison of predicted in-cylinder pressure traces during the compression stroke resulted from the single zone combustion model before and after optimization, along with the

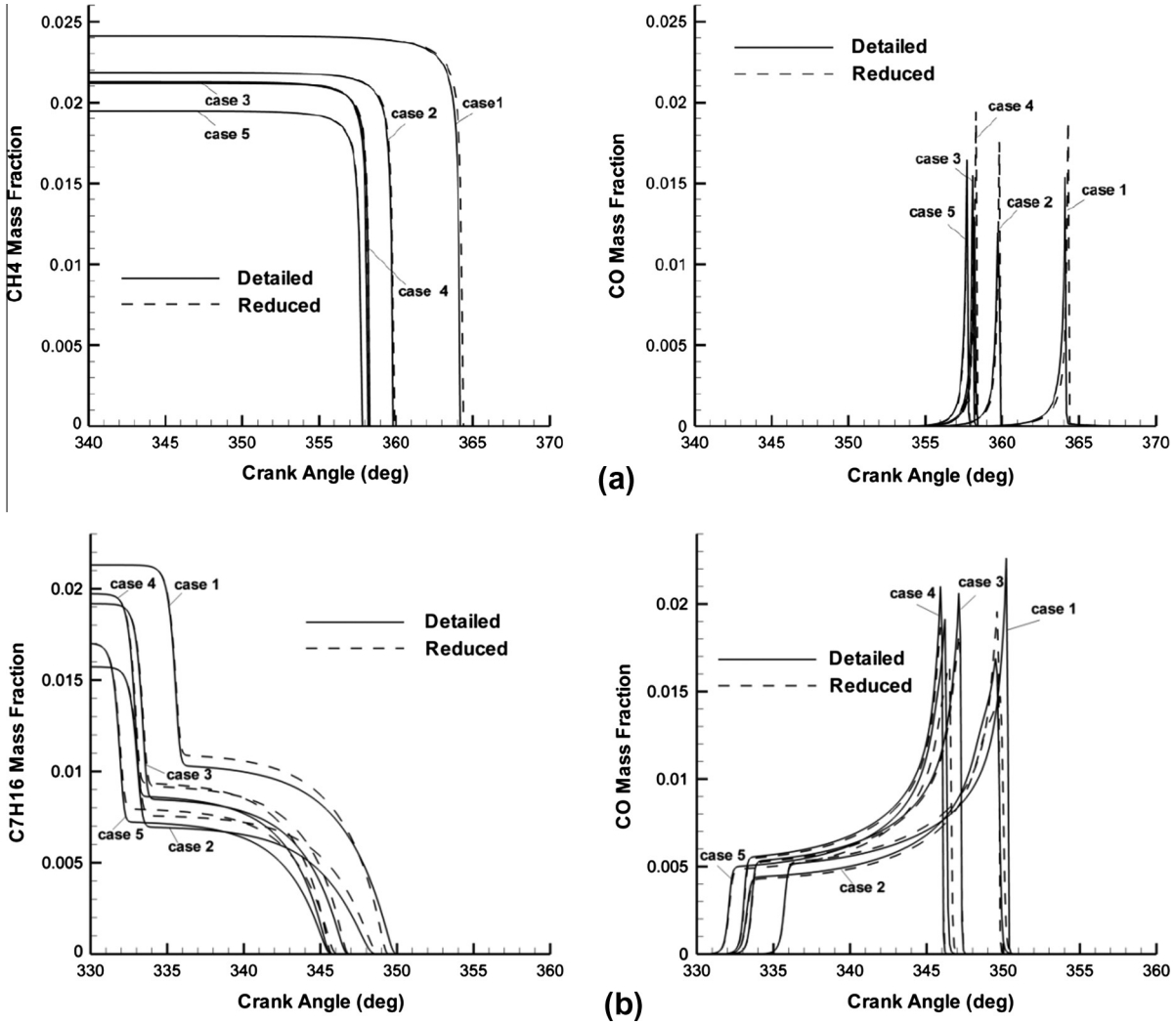

Fig. 10. Comparison of mass fraction for some selected species between (a) the detailed natural gas mechanism and its reduced mechanism generated for case 5 and (b) the detailed n-heptane mechanism and its reduced mechanism generated for case 2.

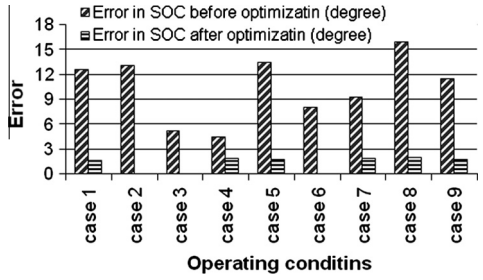

Fig. 11. Error in prediction of SOC for fuel blend of natural gas/n-heptane in HCCI combustion engine for reduced mechanisms relative to experimental ones at all considered cases.

corresponding experimental data for some selected operating conditions mentioned in Table 4. These graphs demonstrate that the ability of the model using optimized mechanism in predicting the in-cylinder pressure in comparison with experimental data through the compression stroke is reasonably well and also it provides accurate prediction of ignition timing. It is well known that the SZCM over-estimates the combustion rates after the ignition period, due to its assumptions. If the rest of the cycle is needed to be analyzed thermal and composition distributions of cylinder charge should be considered. This goal is achieved by employing multi-zone models but much longer simulation time is required in comparison to SZCM. A large number of simulations are required to optimize the mechanism using GA optimization, and the use of multi-zone models is prohibitive. It is observed that SZCM is exceedingly accurate for the purpose of this paper. As a result, in this study, single-zone modeling was adequate for following the auto-ignition process up to the beginning of the main combustion. From this figure it is evident that a significant discrepancy is observed when applying mechanism with the original combustion parameters to the model with the experimental results. Thus, the optimization is necessary. The proposed reduced mechanism for the present study is provided as additional Information with this article.

Fig. 13 compares the predicted pressure trace obtained experimentally with those of using the Golovichev's, Curran's and proposed mechanisms. While both the Golovichev's and Curran's mechanisms exhibit discrepancies in predicting ignition timing, the proposed mechanism provides reasonable agreement with the measurements for predicting the ignition timing.

Table 7 shows the ability of the above mentioned mechanisms in predicting SOC. The proposed mechanism increased the simulation speed by 57 times in comparison with Golovichev's mechanism and 99.9 times in comparison with Curran's mechanism.

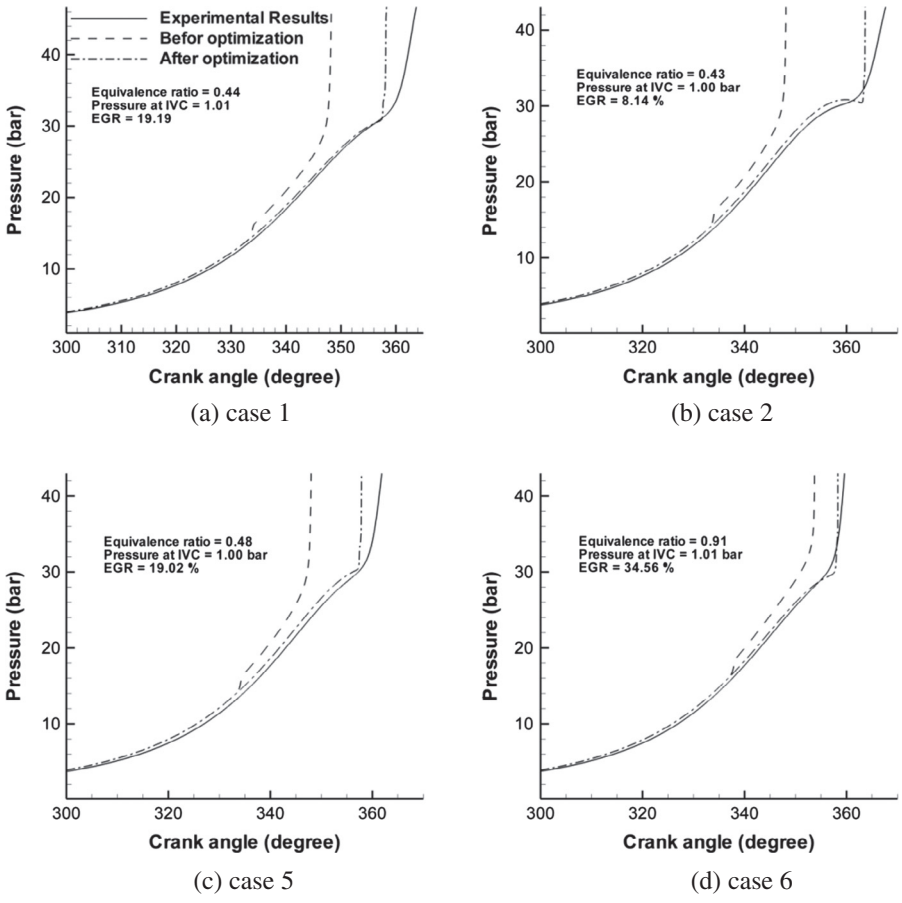

Fig. 12. Comparison of predicted in-cylinder pressure traces during the compression stroke resulting from the single-zone combustion model before and after optimization with the corresponding experimental data.

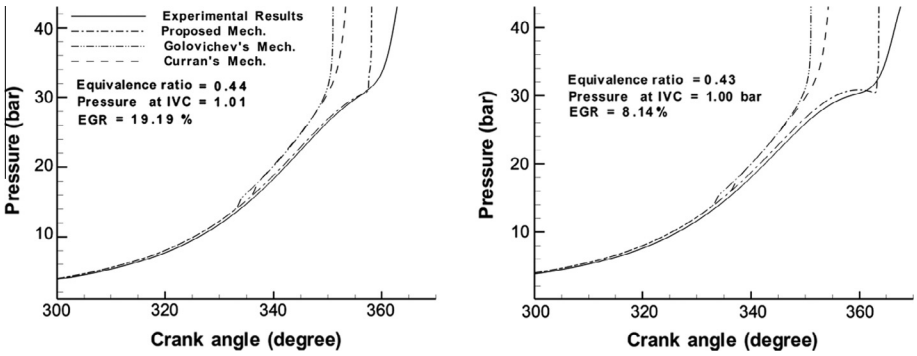

Fig. 13. Comparison of predicted in-cylinder pressure traces resulting from the single-zone combustion model employing the Golovichev's mechanism, the Curran's mechanism, and the proposed mechanism with the corresponding experimental data.

Table 7
Comparison of proposed mechanism, Golovichev's mechanism, and also Curran's mechanism.

mechanism	Species	Reactions	Error in SOC for case 1 (Degree)	Error in SOC for case 2 (Degree)	Simulation time (h:min:s)
Curran	561	2539	10.0	12.0	10:01:21
Golovichev	57	290	10.8	13.6	00:01:24
This work	41	109	1.7	0.1	00:00:36

6. Conclusion

Fuel blending has been recognized as a technique to control combustion timing in the HCCI combustion engine. In the current study, a reduced mechanism of natural-gas/n-heptane fuel with the combination of two developed reduced mechanisms of natural gas and n-heptane fuel, based on the GRI-Mech. 3.0 and Golovichev's mechanisms, was proposed. Reduction procedure to

develop the reduced mechanisms was based on an integrated method that utilizes DRGEP and PCA reduction methods. The mechanism reduction achieved reduction ratios of 64 and 30% for the number of species and also 88 and 67% for the number of reactions (for the natural gas and n-heptane mechanisms respectively). These were achieved within tight error limits for prediction of CA50, peak pressure, and maximum heat release. The combination of the generated reduced mechanisms is used to develop a reaction mechanism for a fuel blend of natural-gas/n-heptane. The combined mechanism was optimized using Genetic Algorithm and validated against experimental data for predicting ignition timing in the natural-gas/n-heptane fuelled HCCI engine.

The proposed reduced mechanism for fuel blend of natural-gas and n-heptane, which contains 41 species and 109 reactions, and the generated reduced mechanisms for natural gas and n-heptane are included as additional information with this article.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.enconman.2013.12.005.

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