

Extension of the Phenomenological 3-Arrhenius Auto-Ignition Model for Six Surrogate Automotive Fuels

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ABSTRACT

An existing three-stage ignition delay model which has seen successful application to Primary Reference Fuels (PRFs) has been extended to six surrogate fuels which constitute potential candidates for future Homogeneous Charge Compression Ignition (HCCI) engines. The fuels include petroleum-derived and oxygenated components and can be divided into low, intermediate and high cetane number groups. A new methodology to obtain the model parameters is presented which relies jointly on simulation and experimental data: in a first step, constant volume adiabatic reactor simulations using chemical kinetic mechanisms are performed to generate ignition delays for a very wide range of conditions, namely variations in equivalence ratio, Exhaust Gas Recirculation (EGR), pressure and temperature. Based on this “virtual shock tube” data the 3-Arrhenius parameters for low- and high-temperature ignition delay and Negative Temperature Coefficient (NTC) regimes are determined by means of a genetic algorithm. In a second step, the parameterized 3-Arrhenius model is assessed by means of experimental auto-ignition delay data from a Rapid Compression Expansion Machine (RCEM). This data revealed a systematic over prediction of the low and high temperature ignition delays. A refinement of the pre-exponential parameters of the low and high ignition delay terms was carried out resulting in excellent agreement over a large range of operating conditions between model and measurements for all six fuels. The approach proposed in this study hence combines 1) the efficient generation of ‘numerical’ ignition delay data for surrogate fuels at different pressure, temperature, equivalence ratio and EGR levels for which the variations span much larger ranges than typically available from experiments, 2) the rapid parameterization of this dataset by means of genetic algorithms to derive “initial estimates” for the three-stage ignition model parameters, for which in 3) the final values are determined by retuning the pre-exponential parameters of the low and high ignition delay terms based on a wide range of measurements in a RCEM. To conclude the new methodology has successfully extended the application of the 3-Arrhenius model to Toluene Reference Fuels (TRFs), longer-chain alkanes and oxygenated fuels. The efficiency and robustness of the new methodology compared to the traditional usage of exclusively experimental data for model parameterization considerably extends its applicability to auto-ignition modeling for a variety of fuels. This will allow for broader use of such models for different fuels for HCCI engine calibration and control.

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INTRODUCTION

Over the past decades oil and transportation industries have witnessed an increasing need to develop more efficient and cleaner engines. Homogeneous Charge Compression Ignition (HCCI) combustion engines hold the potential to achieve both of these goals. The technology seeks to combine advantages of diesel and gasoline engines, i.e. achieving the efficiencies of compression-ignition engines while simultaneously favoring emissions similar to gasoline engines [1]. Unlike diesel or gasoline engines, the onset of combustion is not governed by an explicit ignition-controlling event such as fuel injection

or spark advance. In HCCI combustion, fuel and intake charge are homogeneously mixed, prior to compression and subsequent auto-ignition. At the point of ignition a very quick and violent combustion takes place leading to a rapid increase in pressure and temperature. Due to the homogeneity of the fuel-gas mixture, “volumetric” ignition occurs at various locations in the combustion chamber, leading to no distinct flame propagation [1]. Therefore the onset of ignition becomes challenging to control and to predict.

To address this matter, numerous approaches to model HCCI combustion have been proposed in literature. Zero-dimensional, single-zone models are simple predictive tools and are considered to provide the core for HCCI modeling [1]. Although this approach has found very useful applications in the validation and comparison of chemical reaction mechanisms, in the prediction of HCCI combustion trends (e.g. ignition delays and peak pressures) and remains computationally inexpensive, it suffers from shortcomings due to the assumption of complete homogeneity throughout the entire combustion chamber [2]. Multi-zone models can address the issue of slight stratification in the combustion chamber and such models were successfully applied for the prediction of HCCI combustion by [3], [4]. However, in the presence of more substantial and detailed stratification - e.g. due to incomplete mixing or thermal inhomogeneity due to differences in wall temperature and corresponding heat transfer - multi-zone models are no longer suitable and full 3D-CRFD simulations are necessary [5]. Most often, global (e.g. [6], [7], [8], [9]) to reduced chemical kinetics (e.g. [10], [11], [12], [13]) are integrated into a 3D-CFD code. However, thanks to growing computational power highly detailed chemical kinetics like [14], [15], [16], [17] will also be adopted in the foreseeable future for these complex simulations. Due to the high computational demand several 3D-CFD simulations employ the simpler Reynolds Averaged Navier Stokes (RANS) approach. In many cases, turbulence-chemistry interaction is neglected by carrying out so-called “direct integration (DI)”, see e.g. [11], [18], although more sophisticated approaches using e.g. conditional moment closure (CMC) have also been reported for HCCI [19] and LTC type combustion [20], [21]. However thanks to the continuous growth of computational resources more complex flow field models such as LES and DNS have also been coupled with chemistry to simulate HCCI combustion [22], [23], [24], [25].

Even if 3D-CRFD is a powerful tool for the simulation of HCCI combustion, it important to note that it is *significantly* more costly compared to zero-dimensional models in terms of computational requirements. Therefore they are not ideal to use in engine applications [26], [27], [28]. To address this shortcoming, this paper focuses on phenomenological modeling of HCCI auto-ignition. This approach has been successfully implemented in the past, e.g. in [29], [30], [31], [32], [33], [34], [35], [36], [37], [38]. Due to their simplicity these models can serve as powerful tools for engine development and also in terms of closed loop engine control [39], [40].

Many factors must be taken into consideration to model HCCI auto-ignition. In general it is predominantly influenced by fuel chemistry [41]. Equivalence ratio, EGR rate, compression ratio, intake and in-cylinder temperature, fuel spray characteristics (which influences mixture stratification), heat transfer between gas and engine walls and in-cylinder turbulence (which influences incylinder temperature homogeneity) can also influence auto-ignition [42], [43], [44], [45], [46], [47], [48], [49]. The model presented in this paper covers numerous surrogate fuels, representative of both conventional and alternative fuels with dissimilar auto-ignition characteristics, over a wide operating range. This will help to advance some of the major

challenges of HCCI combustion, i.e. determine which fuels are suitable for HCCI combustion, and achieve full control over the onset of ignition and of combustion phasing [1], [50], [51], [52].

FUELS

Table 1 shows the target automotive fuels and their corresponding surrogate fuels. The fuels’ nomenclature is described in Table 2.

Table 1. Original fuels and their corresponding surrogates. The mixture volume percentages and components are included in the table.

		Low CN (≈38)	Mid CN (≈50-55)		High CN (≈70)
Non-Oxygenated	Original Fuels	57.5% _{Vol} DK B0-1 + 42.5% _{Vol} Eurosuper (CN 39.6)	DK B0-1 (CN 55.6)	DK B0-2 + EHN (CN 55.8)	HVO (CN>74.6)
	Surrogate Fuels	PRF41 (CN 39.6) 41% _{Vol} iso-Octane + 59% _{Vol} n-Heptane	n-Heptane (CN 55.28)	TRF 7.3 (CN 51.42) 92.7% _{Vol} n-Heptane + 7.3% _{Vol} Toluene	n-Decane (CN 76)
Oxygenated	Original Fuels	66.5% _{Vol} DK B0-1 + 33.5% _{Vol} ETBE (CN 36.5) (4.4% _{Mass} O ₂ content)	64% _{Vol} DK B0-1 + 36% _{Vol} RME (CN 55.2) (4.2% _{Mass} O ₂ content)		-
	Surrogate Fuels	BVP 31.8 (CN 37.8) 68.2% _{Vol} n-Heptane + 31.8% _{Vol} n-Butanol (7.67% _{Mass} O ₂ content)	BVP 9.1 (CN 50.05) 90.9% _{Vol} n-Heptane + 9.1% _{Vol} n-Butanol (2.29% _{Mass} O ₂ content)		-

The fuels are divided into three cetane number categories ranging from low to high values and also into non-oxygenated and oxygenated fuels. The surrogates were all chosen to a) roughly match the cetane number (CN) of the original fuel; b) have available chemical reaction mechanisms which describe their pyrolysis/oxidation kinetics and, c) have a boiling temperature below 200°C (the highest is n-decane at 174°C). The latter constraint is limited by the maximum temperature of the RCEM’s wall heating, which serves to evaporate the fuel. The original fuels are all of the full boiling range type, with 100% evaporation at temperatures over 300°C (see Figure 1). Therefore they could not be prepared for HCCI combustion in this RCEM.

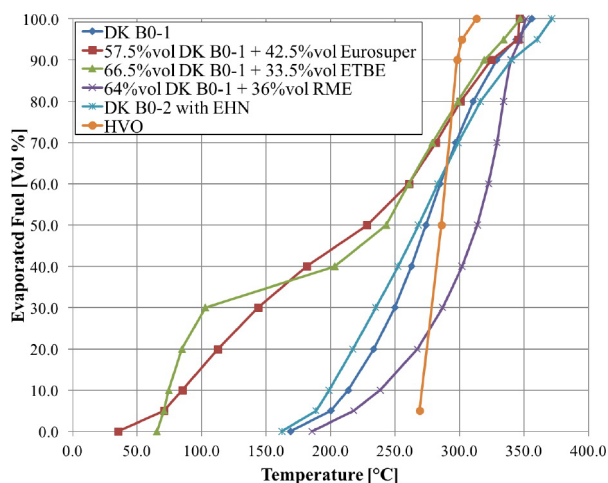


Figure 1. Distillation curve of the original full boiling range fuels.

Table 2. Fuels' nomenclature.

BVP	Butanol volume percentage
DK B0-1	Standard European diesel fuel
DK B0-2	Low quality diesel fuel
EHN	Ethyl-Hexyl-Nitrate
ETBE	Ethyl <i>tert</i> -butyl ether
HVO	Hydrogenated vegetable oil
PRF	Primary reference fuel
RME	Rapeseed methyl ester
TRF	Toluene reference fuel

A number of different ways have been proposed to determine ignition delays from shock-tube and RCEM experimental data, for which discussions and definitions can be found in e.g. [13], [30], [53], [54], [55]. In this paper ignition delays were calculated based on the methodology of Mitakos et al. [54], with the procedure shown in Figure 2. Here the heat release rate was used to determine when low and high temperature ignition delay occurred. Piston position equal to 22mm was chosen as time zero. The choice of this position is selected since it is sufficiently before any LTR ignition of the experimental matrix.

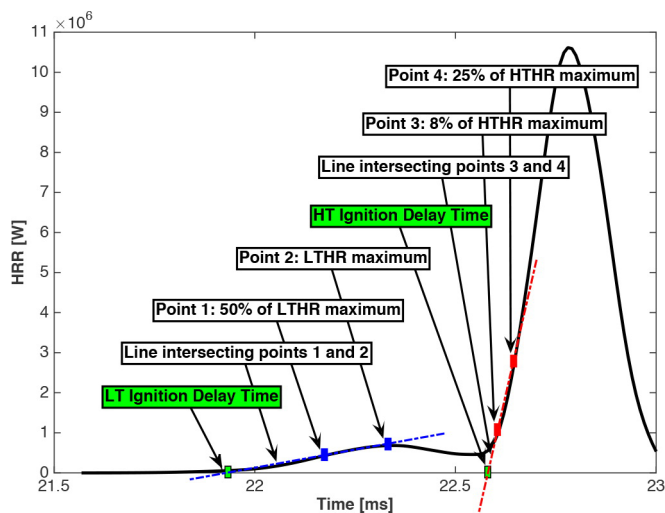
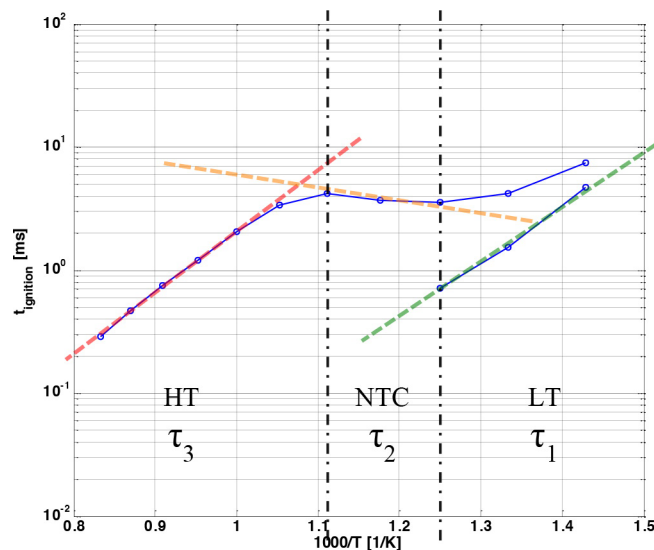


Figure 2. Definition of low and high temperature ignition delays.

AUTO-IGNITION MODEL

The 3-Arrhenius model is a phenomenological auto-ignition tool to predict low and high temperature ignition delays of HCCI combustion processes. Contrary to [29], [30], [31], [32], [33], [34], [34] - which show single Arrhenius expressions with coefficients that are independent from temperature for auto-ignition predictions and can therefore not predict the entire S-shaped auto-ignition curve in Figure 3 - this model can compute ignition delays over the entire auto-ignition temperature range of a fuel, similar to the models [35], [36], [37], [38]. The 3-Arrhenius model was originally proposed by Weisser [39] and further extended by Vandersickel [56] to account for the influence of inert gas on ignition delay, indicated in the term $[inert/O_2]$. For a RCEM or an engine the latter is defined as $(100 - [O_{2,IVC}])/[O_{2,IVC}]$ where $[O_{2,IVC}]$ is equal to the concentration of oxygen at inlet valve closure. In the case where an EGR percentage is given $[O_{2,IVC}] = 21 \cdot (100 - EGR)/100$. In this paper inert gas is referred to as EGR which is composed of 20%_{vol} CO₂ and 80%_{vol} N₂. As the name of the model states, the 3-Arrhenius model comprises three Arrhenius-like ignition terms (shown in logarithmic form in equations (2), (3) and (4)). This enables the model to predict ignition delays over the low temperature τ_1 , negative temperature τ_2 and high temperature τ_3 coefficient domains. Their respective regimes and temperature dependence are indicated exemplary on the Arrhenius ignition delay plot in Figure 3. The coupling of the three ignition terms is shown in equation (1).

Figure 3. An exemplary Arrhenius ignition delay plot. The three domains correspond to the three 3-Arrhenius model terms τ_1 , τ_2 and τ_3 . LT: low temperature; NTC: negative temperature coefficient; HT: high temperature.

The low temperature and the negative temperature coefficient reactions occur sequentially leading to a two-stage ignition path. Therefore they are directly added to one another. The high temperature reactions lead to a parallel single-stage ignition path. This is why it is an independent term. The reference pressure p_{ref} in equations (2), (3) and (4) is equal to 1 bar.

$$\frac{1}{\tau} = \frac{1}{\tau_1 + \tau_2} + \frac{1}{\tau_3}$$

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$$\ln(\tau_1) = \ln(A_1) + \beta_1 \ln\left(\frac{p}{p_{ref}}\right) + \left(\frac{T_{A_1}}{T}\right) \quad (2)$$

$$\begin{aligned} \ln(\tau_2) = & \ln(A_2) + \beta_2 \ln\left(\frac{p}{p_{ref}}\right) + b_2 \ln(T) \\ & + \left(\frac{T_{A_2}}{T}\right) + c_2 \ln(\Phi) \\ & + d_2 \ln\left(\frac{[inert]}{[O_2]}\right) + \left(\frac{[inert]}{[O_2]} \frac{e_2}{T}\right) \end{aligned} \quad (3)$$

$$\begin{aligned} \ln(\tau_3) = & \ln(A_3) + \beta_3 \ln\left(\frac{p}{p_{ref}}\right) + \left(\frac{T_{A_3}}{T}\right) \\ & + d_3 \ln\left(\frac{[inert]}{[O_2]}\right) \end{aligned} \quad (4)$$

The 3-Arrhenius model can be used to predict ignition delays for either constant or transient pressure and temperature conditions by integrating equations (5) and (6) along the pressure/temperature traces, from time zero to the ignition delay time as originally proposed in [57].

$$\int_{t_0}^{t_{LT \text{ ignition}}} \frac{1}{\tau_1} dt = 1 \quad (5)$$

$$\int_{t_0}^{t_{HT \text{ Ignition}}} \frac{1}{\tau} dt = 1 \quad (6)$$

The total number of parameters of the model is 14 (A_i, β_i, T_{A_i} , where $i=1,2,3$; b_2, c_2, d_2, e_2 and d_3). The high dimensionality of the system demands a high number of data points for its calibration and a robust parameterization methodology as will be discussed in the following.

Parameterization Options

The original parameterization process of the 3-Arrhenius model presented by Vandersickel et al. [56], was based on experimental shock-tube data described in [56], [53] and the parameterization was carried out first by performing a sensitivity analysis to determine the model's dependencies on pressure, air-to-fuel ratio and EGR. Then a linear regression was used for the parameterization of the low temperature ignition delay. Finally a non-linear regression was adopted for the negative temperature coefficient and high temperature

terms. This procedure did not make use of an optimization tool hence necessitating considerable efforts by the user. Furthermore at most 45 stock-tube experimental operating conditions per fuel were available for the parameterization.

For the six new fuels investigated in the present study, no shock-tube data was available, therefore the previous parameterization method could not be adopted. However ignition delay data from a rapid compression and expansion machine was available enabling alternative parameterization approaches [58]. To parameterize the 3-Arrhenius model based on transient data, conceptually one could combine a parameter optimization tool, e.g. genetic algorithm, to guess the model's parameters, then feed them into the ignition integral using an experimental pressure/temperature trace and compare the predicted ignition delays to the measured values. This procedure was tested but encountered two difficulties: A) how does one define the initial range of the parameters so that the optimization tool does indeed find a global minimum in a reasonable amount of time and B) the relatively low number of operating conditions which were available for the optimization. With only 18 operating conditions measured for each fuel it was quite probable that the optimization may not converge towards a global minimum, but a local one. Due to these difficulties a new procedure was developed to address the issues mentioned above, namely I) how to provide data that allows one to make an initial guess of the 3-Arrhenius parameters for the optimization tool; II) how to speed up optimization time while maintaining a good final result and III) provide enough optimization data to allow for convergence towards a global and not local minimum. The approach that followed resorts to constant volume ignition delay data for a wide range of operating conditions for the optimization. This also eliminates the need to use an ignition integral, which (as mentioned above) together with a genetic algorithm for parameter optimization is time consuming. In the absence of experimental shock-tube data, perfectly stirred reactor (PSR) calculations have been carried out using detailed chemical reaction mechanisms for all six new fuels. Given the low computational expense, a large number of "virtual shock-tube" operating conditions can be generated, which serve as a thorough optimization database to determine initial estimates for the fourteen 3-Arrhenius parameters.

Since the optimization was based on simulated data and not measurements there was still a need to validate the final parameters. This is where the experimental data from Bardi et al. [58] is utilized to re-tune the model.

0D PSR SHOCK-TUBE SIMULATIONS

The first step of the methodology for the parameterization of the 3-Arrhenius model consists of generating ignition delay data for the parameterization by means of 0D adiabatic constant volume PSR calculations using CHEMKIN. The chemical reaction mechanisms which were used for the simulations are listed in Table 3 and exhibit varying degrees of complexity.

Table 3. Chemical reaction mechanisms used for the simulated shock-tube. FR: flow reactor; ICE: internal combustion engine; JSR: jet-stirred reactor; LF: laminar flame; OFDF: opposed flow diffusion flame; PF: premixed flame; RCM: rapid compression machine; SR1: static reactor; SR2: stirred reactor; ST: shock-tube.

Fuel & CN	Reaction Mechanism	#Species	#Reactions	Validated With
<i>n</i> -Heptane CN55.3	Wang [10]	73	296	PF, JSR, ST, ICE
PRF41 CN39.6	Wang [10]	73	296	PF, JSR, ST, ICE
TRF7.3 CN51.4	LLNL 1 [17]	1550	5935	RCM, JSR, ST
BVP9.1 CN50.1	Saisirirat [16]	1046	4398	ICE, JSR
BVP31.8 CN37.8	Saisirirat [16]	1046	4398	ICE, JSR
<i>n</i> -Decane CN76.0	LLNL 2 [15]	2116	8130	ST, FR, RCM, SR1, SR2, LF, OFDF, ICE,

It is important to note that the mechanism of Wang et al. [10] is a reduced version of the detailed gasoline surrogate mechanism of Mehl et al. [17] with 1389 species and 5935 reactions. The reduced mechanism's performance is very similar to the detailed mechanism, albeit at a much lower computational cost. Therefore the former was chosen over the latter for the simulation of *n*heptane and PRF.

In total 250 operating conditions for each fuel were simulated, with sweeps in pressure, air-to-fuel ratio and inert gas (EGR); the respective ranges are given in Table 4.

Table 4. Simulated shock-tube operating conditions.

Simulated Shock-Tube Operating conditions	
p	20, 30, 40, 50 bar
ϕ	0.33, 0.5, 1
T	700 – 1200 K with 50K resolution
EGR	0, 20, 30, 40%

To keep the range as broad as possible, a wide spread of operating conditions was chosen. The resolution of pressure, ϕ , temperature and EGR was chosen so that each variable could clearly show its influence on ignition delay. A fine resolution of 50 K in initial temperature was simulated, so that the s-shaped ignition curve (see e.g. Figure 3) had enough data points for the optimization. This is of particular importance in the NTC region where the slope changes from positive to negative. Even finer resolutions in temperature (25 K) were also assessed, however the differences in the R^2 and $RMSE$ of predicted ignition delays were negligible. The validation range of the chemical reaction mechanisms was also taken into consideration, so that their validated range was part of the simulated conditions. Furthermore the computational demand of the 0D shock-tube

simulations was quite low. All 250 operating conditions with the most detailed mechanism (LLNL 2 [15]) took roughly 62 hours on one CPU. The most reduced mechanism (Wang [10]) terminated in around 20 minutes.

Ignition delays were determined from the simulated HRR in the same manner as in the RCEM [54], for which the methodology is outlined in Figure 2. This approach is valid thanks to the similarities between RCEM and shock-tube ignition delay data, highlighted in the works of Mitakos [54] and Vandersickel [56]. Furthermore using the same methodology ensured a consistent definition of ignition delay throughout the entire parameterization process, i.e. both for simulated shock-tube and RCEM.

MODEL PARAMETERIZATION USING A GENETIC ALGORITHM

With the database of simulated ignition delays now ready, the next step was to provide an initial guess of the 14 parameters for the GA input. Due to the high dimension of the optimization and also the large range of possible values which the parameters can take, the better the initial guess, the easier it is for the GA to find a global minimum. To find a good initial guess the three terms of the 3-Arrhenius model were linearly regressed onto their corresponding domains of the Arrhenius curve as shown in Figure 3. Since the low temperature term only has three parameters and can be regressed onto a straight line, the corresponding values from the linear regression were considered final and hence were not modified by the GA. The validity of this assumption will be discussed in the results section. Furthermore it is important to note that this reduces the dimension of the optimization from 14 to 11, which helps to reduce computational time and can improve convergence.

The fitness functions of the genetic algorithm were chosen to be the R^2 and $RMSE$ of low and high temperature ignition delay. The latter is logarithmically weighted to balance the importance of all ignition delays over the entire temperature domain. This is important since the s-shaped ignition delay curve can cover multiple orders of magnitude for the given temperature range (e.g. see Figure 3). Therefore without the logarithmic balancing, discrepancies between modeled and simulated shock-tube ignition delays at the lower temperature domain would result in higher variations in $RMSE$ than the high temperature domain.

RCEM EXPERIMENTAL DATA

The experimental data for validation of the model's parameters were taken from Bardi et al. [58]. The employed rapid compression expansion machine allows for accurate control of the boundary conditions and - in addition to pressure indication - features optical access through a large quartz window in the piston allowing for high-speed intensified imaging. Excellent optical access is provided together with the independent setting of operating parameters, such as compression ratio, charge initial pressure and temperature. A transparent virtually flat piston with a bowl depth of 2 mm was utilized. A fully heated combustion chamber provided a homogeneous, steady-state, pre-compression temperature distribution. A very good temperature pre-compression homogeneity