

Characterizing Gaseous Fuels for Their Knock Resistance based on the Chemical and Physical Properties of the Fuel

Sander Gersen, Martijn van Essen, Howard Levinsky, and Gerco van Dijk
DNV GL Oil & Gas

ABSTRACT

A method is described to characterize the effects of changes in the composition of gaseous fuels on engine knock by computing the autoignition process during the compression and burn periods of the engine cycle. To account for the effects of fuel composition on the in-cylinder pressure and temperature history relevant for knocking, changes in heat capacity of the air-fuel mixture and in the phasing of the combustion process are also incorporated in the method. Comparison between pressure profiles measured in a lean-burn, high-speed medium-BMEP gas engine and the calculated pressure profiles at non-knocking conditions shows that the method accurately computes the in-cylinder pressure history when varying the fuel composition. To characterize gaseous fuels for their resistance to knock, a propane-based scale (Propane Knock Index, PKI) is reported in this study. On this scale, the knock resistance for a given gaseous fuel mixture is expressed as an equivalent fraction of propane in methane under identical engine conditions. The chemical mechanism used to compute the autoignition delay time is optimized based on RCM measurements performed at engine conditions. After optimization, the knock resistance as measured by the knock-limited spark timing (KLST) and calculated using PKI was found to agree within the uncertainty of the measurements (± 0.75 °CA) for all fuels studied, including hydrocarbon mixtures representative for liquefied natural gas (LNG) and natural gas/H₂/CO mixtures. In contrast to the method reported here, comparison of KLST for the range of gases studied with methane numbers calculated using the AVL and the MWM methods points to shortcomings in these yardsticks.

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INTRODUCTION

Increasingly stringent regulations regarding CO₂ emissions, depletion of local fuel supplies and the growing desire to enhance the sustainability of the energy supply have resulted in an increasing diversity of pipeline natural gases, a growing interest in using (bio-) LNG and CNG as a transportation fuel and the (possible) introduction of hydrogen containing gases into the natural gas infrastructure. As a result, gases with significantly different composition can be or will be traded and distributed. These variations in the gas composition influence the so-called knock resistance of the fuel. Mild engine knock increases pollutant emissions and can damage the engine in the long term, while severe knock can physically destroy the engine in seconds. Knock-protection systems safeguard against sudden physical damage, but at the expense of lower fuel efficiency and/or lower power output, or even disrupted engine service. Thus, engine knock should be avoided [1]. To ensure that the gas-fueled engines used in, for example, cars, trucks, ships and CHP units are matched with the expected variations in fuel composition, the knock resistance of the fuel must be characterized, and subsequently specified unambiguously.

The occurrence of engine knock is caused by autoignition of unburned fuel mixture, the so-called end gas, in the cylinder before the mixture is completely consumed by the propagating flame. Several empirical methods, such as the methane number approach

[2], have been developed to classify gaseous fuels for their knock sensitivity. These traditionally used methods, in which the knock resistance of a given fuel is compared with an equivalent methane/hydrogen mixture, suffer from a number of shortcomings [3, 4]. Perhaps most apparent of these is that the number is attached to the behavior of an individual engine under stoichiometric conditions, while the actual diversity of installed designs is great, and modern machines tend to operate substantially fuel lean and at higher pressures. In addition, the suitability of hydrogen as a reference gas has been disputed [3, 5] as has been the method of accounting for butane and higher hydrocarbons [6]. Furthermore, a methodology that provides insight into the origins of knock in engines by describing the microscopic details of the effects of fuel composition on cylinder processes is particularly valuable for designers and operators who wish to optimize power output while maintaining knock-free operation. As such, the necessity of having a physically correct and unambiguous method to characterize the knock resistance of the variety of fuels for the variety of engine types in the market has become urgent in recent years.

Autoignition of the unburned end gas in gas engines is initiated by rapid (exothermic) oxidation reactions, leading to heat release that exponentially increases the temperature, causing the overall reaction to auto-accelerate. Experimental studies performed in RCMs and

shock tubes show that the autoignition behavior of fuels depends strongly upon the composition, temperature (T), pressure (P) and stoichiometry [e.g., 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20]. For example, the autoignition delay time of methane-based fuels decreases substantially with increasing pressure and temperature, and with increasing fractions of hydrogen and higher hydrocarbons, such as ethane, propane and butane in the fuel. Furthermore, the autoignition promoting effect of these components can strongly depend upon the temperature. A detailed analysis of the origins of the occurrence of engine knock therefore requires knowledge of the time-dependent state (P , T) of the unburned end gas as well as any changes in this state caused by varying composition. The pressure and temperature of the end gas vary with time during the engine cycle due to the combination of mechanical compression from piston motion and compression from the progressive combustion of the charge, related to the so-called combustion phasing. Changes in the gas composition can lead to substantial variations in the thermodynamic properties of the fuel mixture [3] and in the phasing of the combustion process [21, 22, 23], which result in changes in the temperature and pressure of the end gas. For example, in [22] it has been shown that the addition of hydrogen to natural gas increases the burning velocity (combustion phasing) and reduces the heat capacity. Together, these two effects result in a substantial increase in the temperature and pressure of the end gas and is the major cause of the reduced knock resistance with increasing fractions hydrogen in natural gas. Thus, to quantify the progress of autoignition under engine conditions for different gas compositions, this composition dependence of the variation in the temperature and pressure of the end gas must also be quantified.

In this paper, we present a method to characterize the knock resistance gaseous fuels based on the combustion properties of the fuel mixtures themselves. In this method, engine knock is predicted by modeling autoignition of the compressed end gas during the engine cycle, including the changes in in-cylinder pressure and temperature experienced by the end gas upon changing gas composition [21], as described above. The model is demonstrated for a constant-speed, medium-BMEP engine for a wide range of fuel compositions typical for natural gases, containing varying fractions of methane, ethane, propane and butane. In addition, the success of the method to account correctly for the effects of CO_2 , N_2 and H_2 in natural gas is demonstrated.

EXPERIMENTAL WORK

Gas Engine

A lean-burn, high-speed, turbo-charged, intercooled 6-cylinder gas engine (MAN) for combined-heat-and-power applications (CHP) was used for the engine measurements. Table 1 lists the key specifications of the engine. We remark here that this CHP system maintains constant fuel/air ratio, engine speed and power output. The engine management systems and additional instrumentation provided precise adjustment, monitoring and registration of all relevant engine settings and process conditions. The in-cylinder pressures in all cylinders were measured with Kistler type 6052 piezoelectric pressure sensors and a Kistler type 5011 charge amplifiers connected to a Smetec Combi-Pro indication system. A crankshaft-mounted pulse generator provided 0.1 °CA resolution for the cylinder pressure data

acquisition. The Smetec Combi-Pro system was also used for knock detection through monitoring of the maximum amplitude of pressure oscillations in the high-pass-filtered cylinder pressure data in a window of consecutive cycles. Threshold settings used were 2 bar for the amplitude and 30 cycles for the window. For knock-limited spark timing (KLST) experiments ([3], see below), the knock limit was chosen to be one knock event or cluster of knock events within a 15 minute steady-state test run.

Table 1. Key specifications gas engine

Engine make & type	MAN E2876LE302
Engine configuration	- 6 cylinder in-line - turbocharged - intercooled
Combustion system & combustion stoichiometry	- mono-gas - open chamber - lean-burn
Rated power & speed (corresponding BMEP)	208 kW at 1500 rpm (13.0 bar)
Bore x Stroke	128 x 166 mm
Compression ratio	11.0:1
Engine speed	1500 rpm
Excess air ratio (λ)	1.5
Intake manifold pressure	2.2 bara
Intake manifold temperature	340 K

During the experiments, fuel for the test engine was delivered by a gas mixing unit. The fuel compositions used in the experiments are given in Appendices A and B. All mixture compositions were verified by gas chromatography. Once the composition of the fuel/air mixtures at $\lambda=1.5$ and the other relevant cylinder parameters are set, the pressure traces of the gases under non-knocking condition, needed to test the two-zone thermodynamic model discussed below, are measured at a constant spark timing of 14° CA Before Top Dead Center (BTDC). Subsequently, the spark timing was gradually advanced up to the point of the onset of borderline knock to determine the Knock Limited Spark Timing (KLST).

Rapid Compression Machine

To assess the chemical mechanisms that form the heart of the autoignition part of the model, autoignition measurements in a Rapid Compression Machine (RCM) are reported for species that specifically warranted closer examination (see below). The composition of the fuel-air mixtures for $\lambda=2$ are given in Table 2. The measurements have been performed at temperatures and pressures relevant to the unburned end gas conditions in the engine; pressures (P_c) and temperatures (T_c) after compression ranging from 30-80 bar and 940-1010 K, respectively. As will be seen below, particularly important are the measurements above ~40 bar. All measurements have been compared with numerical simulations in which the effects of heat loss and varying specific volume that occur during the RCM experiments are taken into account. The simulations are performed by using the mechanism developed in [12, 13, 14, 15]. The experimental and simulation procedures are described in detail in [10, 11].

Table 2. Fuel/air mixture compositions in RCM measurements, expressed in mole fraction for $\lambda=2$.

	CH ₄	C ₂ H ₆	C ₃ H ₈	n-C ₄ H ₁₀	i-C ₄ H ₁₀	O ₂	N ₂	Ar
A	0.0476	0	0	0	0	0.1905	0.3	0.4619
B	0.0333	0.0083	0	0	0	0.1917	0.3	0.4667
C	0.0375	0	0.0042	0	0	0.1917	0.3	0.4666
D	0.0397	0	0	0.0025	0	0.1916	0.3	0.4662
E	0.0397	0	0	0	0.0025	0.1916	0.3	0.4662

Description Two-Zone Thermodynamic Combustion Model

As discussed above, variations in gas composition can cause substantial changes in the in-cylinder conditions (P , T) affecting autoignition in the end gas [3, 22, 23]. To predict the effects of the gas composition on the in-cylinder conditions, we apply a two-zone thermodynamic model [21]. In this one dimensional model, the combustion chamber is divided into a zone consisting of unburned fuel-air mixture and a zone with the burned gases, separated by an infinitely sharp flame front. In the model we assume that during the entire engine cycle no heat exchange occurs between the two zones and that the pressure is uniform throughout the cylinder. All gases are considered as ideal gases and the composition of the individual zones is homogeneous. The enthalpy changes of the gases flowing in and flowing out of the cylinder are neglected in the model. The input data needed for the model are the engine geometry, in-cylinder conditions (P , T) at valve closure, operating conditions such as spark timing, air factor, humidity and the fuel composition. The model takes into account that the power output and the fuel/air ratio of the engine are kept constant by a control system that changes the fuel and fuel/air input accordingly.

The time-dependent state of the two zones in the combustion chamber is described using the conservation equations for mass and energy, and the ideal gas equation of state. The mixture is simulated as a single zone until the start of combustion (the spark timing); immediately after spark ignition the burned zone is created and the mixture consists of two zones; a burned zone and an unburned zone (the end gas). After the mixture is completely burned the mixture is simulated again as a single zone. The actual total volume V_T of the cylinder, which is calculated from the geometrical properties of the engine [1], is conserved at any time in the cycle:

$$\frac{dV_T}{dt} = \frac{dV_u}{dt} + \frac{dV_b}{dt}, \quad (1)$$

where the subscript u refers to the unburned mixture and b to the burned gases. By neglecting valve leakage and blowby, we may assume conservation of mass m during the combustion cycle:

$$\frac{dm_u}{dt} = -\frac{dm_b}{dt} \quad (2)$$

The conservation of energy for the unburned and burned zone are written as follows,

$$C_{p,u} m_u \frac{dT_u}{dt} + h_u \frac{dm_u}{dt} = -\frac{dQ_u}{dt} + V_u \frac{dP}{dt} + h_u \frac{dm}{dt} \quad (3)$$

$$C_{p,b} m_b \frac{dT_b}{dt} + h_b \frac{dm_b}{dt} = V_b \frac{dP}{dt} - \frac{dQ_b}{dt} - h_u \frac{dm_u}{dt} \quad (4)$$

where C_p is the heat capacity, h the specific enthalpy, P the pressure and Q the heat transferred to the surrounding surfaces (walls, and piston- and cylinder head). The heat loss Q in both zones is given by

$$\frac{dQ}{dt} = h_c A (T - T_w), \quad (5)$$

where the heat transfer coefficient h_c for both zones is determined by using the Woschni correlation formula [24, 25]. For ideal gases, assuming the pressure is uniform inside the cylinder we can write,

$$P = \frac{m_u R_u T_u}{V_u} = \frac{m_b R_b T_b}{V_b}. \quad (6)$$

The heat capacity and the specific enthalpy of the burned gas and the unburned end gas are calculated as a function of temperature during the combustion cycle using the method proposed in [26] with the thermodynamic data from [12, 13, 14, 15]. The mixture composition of the burned gases, needed as input for the thermodynamic calculations are obtained by using the PREMIX code [27] from the CHEMKIN II suite [26], using the USC Mech Version II reaction mechanism [28]. The composition of the burned gases is calculated numerically at the moment of ignition and is assumed to be constant from the start of the combustion until the exhaust valve opening.

Burning-Rate Model

To solve the governing equations given above we need to find an expression for the mass burning rate (dm_b/dt) that takes into account the operating conditions (e.g. changes in composition) and the engine parameters (e.g. turbulence intensity). The exact prediction of this burning rate is challenging, since the flame propagation is influenced by turbulence in the combustion chamber. To avoid complex CFD calculations, a one-dimensional model is chosen as proposed by Chmela [29] and applied by others [30, 31] in which the rate at which the burned mass is formed is related to the laminar burning velocity (S_L) and a fit parameter C_{model} that accounts for the impact of turbulence;

$$\frac{dm_b}{dt} = C_{model} (1 - x_B) \rho_u S_L t^2, \quad (7)$$

The laminar burning velocity S_L is calculated using the following power-law correlation [1].

$$S_L = S_{L0} \left(\frac{T}{T_0} \right)^\alpha \left(\frac{P}{P_0} \right)^\beta \quad (8)$$

Where S_{L0} is the laminar burning velocity of the fuel/air mixture at the time of spark ignition, also calculated using the PREMIX code as described above for the burned gas composition. The parameters α and β , which take into account the temperature and pressure dependence of laminar burning velocity, are dependent on the fuel composition. These parameters are taken from a database generated for this purpose from numerical laminar burning velocities for a wide range of gas compositions and at operating conditions (pressure, temperature and fuel to air ratio, $\lambda=1.5$) relevant for the gas engine used here. Using this method, only one laminar burning velocity has to be computed in detail for each phasing simulation (at the spark timing), which considerably shortens the computation time needed for the model.

It is known that differential molecular transport effects, as described by the Lewis number (ratio of thermal diffusion and mass diffusion) that is related to stretch effects in laminar flames, affect the combustion behavior of turbulent premixed flames [32, 33] at (mainly) fuel-lean conditions. In [33], it is shown experimentally that there is a strong correlation between the ratio of the turbulent- and laminar burning velocity (S_T/S_L) and the Lewis number for hydrocarbon gas compositions studied. This result emphasizes the importance of accounting for the effects of differential molecular transport of the fuel composition when scaling the laminar and turbulent flame speed. Following the suggestion of [32], we included the Lewis number in Eq. (7), yielding:

$$\frac{dm_b}{dt} = \frac{C_{model}}{Le_{eff}} (1 - x_B) \rho_u S_L t^2, \quad (9)$$

where Le_{eff} is the effective Lewis number, which corresponds to a weighted average of the Lewis numbers of the fuels present in the mixture. The effective Lewis number is calculated following the procedure described in [34, 35].

The set of differential equations (1), (2), (3), (4), (5) and (9) is solved using a fourth-order Runge-Kutta method. The changes in thermophysical properties with changing fuel composition, essential for the correct prediction of changes in the temperature history, are included in the solution of the conservation equations.

RESULTS AND DISCUSSION

Two-Zone Combustion Model

The fit parameter (C_{model}) introduced in the phasing model (Equation 9) to account for the impact of turbulence in the cylinder on the burning rate is determined via a fitting routine by using the pressure trace of the measured methane/air mixture in the engine. As was shown previously [21], a single value of C_{model} , independent of the gas composition, is sufficient to predict the pressure profiles accurately. Based on the fit parameter, engine data and gas composition, the pressure traces of the other gas mixtures (Appendices A and B) have been predicted using the two-zone combustion model. An example of the comparison between the predicted and measured cylinder pressure profiles is presented in Figure 1.

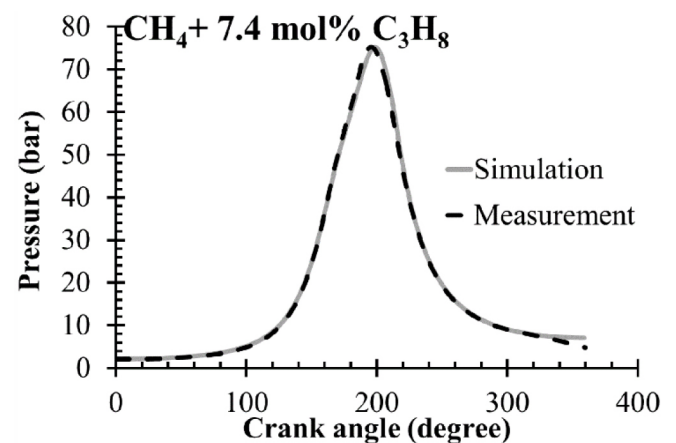


Figure 1. Comparison of measured and simulated non-knocking pressure trace.

Furthermore, Figure 2 shows the comparison of the simulation results with the measurements for a number of fuel mixtures, expanding the scale to emphasize the comparison of the peak pressure; at pressures below 40 bar the measured and simulated pressure traces are identical as shown in Figure 1. For all of the fuel mixtures studied, the maximum deviation in peak pressure is 2 bar corresponding to a deviation in peak temperature of 3 K. As illustrated in Figures 1 and 2, a small systematic shift in the peak pressure is observed in the traces along the crank angle axis in all simulations. Autoignition computations, discussed in detail below, using the experimental pressure traces show negligible differences from those using the predicted pressure trace for the gases studied, demonstrating that the predictive power of the phasing model is adequate for predicting knock.

Here we remark that computations of the pressure profiles without taking the Lewis number into account (taking $Le_{eff} = 1$), resulted in a substantial and systematic underprediction with hydrogen containing gases, up to 5 bar, while the presence of higher hydrocarbons resulted in a systematic overprediction up to 2.5 bar. The introduction of the Lewis number in equation (9) brings the predicted and measured pressure traces into the agreement shown in Figure 2. The Lewis number decreases with increasing fractions of higher hydrocarbons in the mixture, which depresses the mass burning rate and, as a consequence lowers the peak pressure, while a decrease in the Lewis