

# Syngas Production in Piston Engines – Operating Conditions Proposed by Numerical Optimization

H. Gossler<sup>1</sup>, O. Deutschmann<sup>1,2,\*</sup>

<sup>1</sup>Institute for Chemical Technology and Polymer Chemistry, Karlsruhe Institute of Technology, Engesserstr. 18, D-76128 Karlsruhe, Germany

<sup>2</sup>Institute of Catalysis Research and Technology, Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, D-76344 Eggenstein-Leopoldshafen, Germany

## Abstract

An HCCI process is modeled by a single-zone variable volume reactor using elementary-step reaction mechanisms. Numerical optimization is applied to maximize syngas yield at the end of the expansion stroke by varying the initial temperature, initial pressure and equivalence ratio for a fixed engine speed. Suitable constraints were defined, including peak pressure and ignition timing. From these results, maximum syngas yield profiles and the associated operating parameter profiles as functions of engine speed were obtained. Additionally, the same optimizations were done using argon-enriched air in order to attain elevated temperatures.

## Introduction

Piston engines can be viewed as highly dynamic high temperature/pressure reactors and therefore possibly be an attractive alternative to conventional reactors for certain chemical synthesis applications. Depending on the operating parameters, it may be possible to attain reaction conditions that can hardly be achieved in conventional reactors, e.g. the ability to quench the reaction. Additionally, piston engines can be operated very flexibly and react promptly to changes in controls. This poses another advantage that can possibly also be exploited for chemical processes.

Internal combustion engines (ICE) have previously not often been the target of research regarding their use for the production of chemicals. Modeling work concerning ICEs has therefore essentially been done to investigate operating characteristics under conventional conditions, i.e. to produce mechanical energy. Very few studies have dealt with their usage as a reactor, e.g. [1–5]. In these publications, the production of syngas by partial oxidation, i.e. under fuel-rich conditions, of hydrocarbon fuels was studied. In particular, methane and natural gas as fuels were examined. In conclusion, the research which has been conducted so far represents a number of feasibility studies. The experimental results show that the production of synthesis gas via partial oxidation of methane in the most common engine types is possible. In addition, many of these reports state that the engine was also capable of simultaneously delivering power to the shaft. However, it took some effort to ensure stable engine operation in the experiments.

In this study, we further investigate the production of syngas under uncatalyzed partial oxidation and HCCI conditions in an internal combustion engine fueled by natural gas. We apply numerical optimization to propose optimal operating conditions for the production of syngas. This is advantageous since the possible combinations of engine operating parameters that influence the outcome of reactions are abundant. An analysis of

syngas production solely by trial-and-error methods or empirical experiments is therefore impractical.

A computer code (DETCHEM<sup>ENGINE</sup> [6]) was developed, which calculates time-dependent concentration profiles for the compression and expansion strokes of a four-stroke engine using detailed gas phase reaction mechanisms. Syngas yields were derived from these profiles and set as the objective function to be maximized in the optimization problem. The underlying single-zone model corresponds to an idealized batch reactor with a variable volume profile. Due to the assumption of absence of concentration gradients during compression, the model resembles an engine running under HCCI (homogeneous charge compression ignition) conditions. Heat loss is accounted for by using the heat transfer correlation from Assanis et al. [7], which is a modified version of the widely used Woschni heat transfer correlation but tailored to HCCI operating conditions.

Powell's COBYLA optimization algorithm [8], as implemented in the NLOpt library [9], was used to vary initial temperature, initial pressure and equivalence ratio for a given engine speed (up to 6000 rpm). Suitable constraints were defined, including peak pressure and ignition timing. Additionally, lower and upper bounds for each of the optimization variables were set. From these results, maximum syngas yield profiles and the associated operating parameter profiles as functions of engine speed were obtained. The profiles were calculated for a set of different upper temperature bounds, varying from 473 K to 773 K.

## Modeling Approach

The model implemented in the newly developed code DETCHEM<sup>ENGINE</sup> corresponds to an idealized batch reactor with a variable volume profile. It accounts for the temporal change of species concentrations, volume, temperature and pressure inside the combustion chamber. The combustion chamber is considered to be a single zone without spatial variations in concentration

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\* Corresponding author: olaf.deutschmann@kit.edu  
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and temperature. Since a homogeneous mixture is injected into the combustion chamber and selfignites without external influence, the model is expected to be a well-suited approximation of an HCCI engine with a four-stroke operating cycle. Heat loss due to engine cooling is accounted for by using a modified version of the widely used Woschni heat transfer correlation but tailored to HCCI operating conditions [7,10]. Only gas phase reactions are considered; surface reactions are neglected. Furthermore, the gas is treated as an ideal gas.

The simulation begins with a prefilled cylinder with a piston position at bottom center (BC), equivalent to a crank angle of  $180^\circ$ . A full virtual rotation of the crankshaft is then carried out. The compression and expansion strokes are mimicked by a time-dependent volume profile, which is derived from geometrical parameters of the engine according to [11]

$$v(t) = v_c \left( 1 + \frac{1}{2} (r_c - 1) \left[ R + 1 - \sqrt{R^2 - \sin^2 \theta} - \cos \theta \right] \right). \quad (1)$$

In this equation,  $v$  is the instantaneous volume,  $v_c$  is the clearance volume,  $r_c$  is the compression ratio,  $R$  is the connecting rod to crank radius ratio and  $\theta$  is the crank angle, itself a function of time and related to the engine speed  $N$ :

$$\theta(t) = 2\pi N \cdot t + \pi. \quad (2)$$

Only a single cylinder is accounted for and the other two strokes (intake and exhaust) are not considered. The simulation ends at the end of the expansion stroke at BC.

The chemical conversion is modeled by detailed reaction mechanisms. While the explicit description of each elementary step comes at the expense of long computational times, detailed reaction mechanisms allow for a reliable extrapolation to various conditions [12]. Since the primary concern in this paper is the chemical conversion inside the cylinder (not, say, accurate reproduction of pressure profiles), simple global rate expressions cannot be used reliably.

All simulations were carried out using the ‘‘GRI-Mech 3.0’’ reaction mechanism [13]. The mechanism is widely known and is used to model the combustion of natural gas under various conditions. It considers 53 gas-phase species up to  $C_3$  and includes 325 reactions. The mechanism was optimized for methane and natural gas as fuel in the temperature range of 1000 K to 2500 K ( $727^\circ\text{C}$  to  $2227^\circ\text{C}$ ), for pressures up to 10 atm and equivalence ratios between 0.1 and 5.0 in premixed systems. Since the optimization was done primarily with natural gas combustion in mind, it is recommended that the mechanism should not be used to simulate fuels with a main component other than methane (e.g. ethane, propane, methanol, ethylene and acetylene) [13,14].

The Woschni heat transfer correlation [10] has been frequently used to account for heat loss through the cylinder walls in HCCI studies. However, the correlation was developed in particular for diesel engines, whose conditions differ significantly from those in

HCCI engines. Therefore, the heat transfer correlation by Assanis et al. [7] was used in this study. This correlation provides an instantaneous spatially-averaged heat transfer coefficient. It is a modified version of the Woschni heat transfer correlation and provides a spatially averaged value for the cylinder (global heat transfer model). For a detailed insight, the reader is referred to the original publication [7]. The equation is

$$h(t) = \alpha \cdot L(t)^{-0.2} P(t)^{0.8} T(t)^{-0.73} w(t)^{0.8} \quad (3)$$

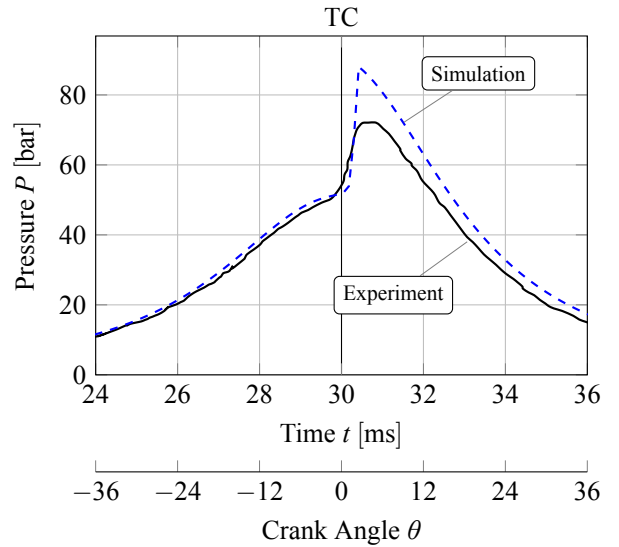
where  $\alpha$  is a scaling factor to match a specific engine geometry,  $L$  is the instantaneous chamber height,  $P$  is the instantaneous pressure (in bar),  $T$  the instantaneous temperature and  $w$  is the instantaneous gas velocity:

$$w(t) = C_1 c_m + \frac{C_2}{6} \frac{v_d T_{\text{ref}}}{P_{\text{ref}} v_{\text{ref}}} (P(t) - P_{\text{mot}}(t)) \quad (4)$$

Here,  $C_1$  and  $C_2$  are constants,  $c_m$  is the mean piston speed,  $v_d$  is the displaced volume,  $T_{\text{ref}}$ ,  $P_{\text{ref}}$ ,  $v_{\text{ref}}$  are reference values of temperature, pressure and volume for which their corresponding values at the beginning of the simulation are used. The last term in the above equation is the difference between the instantaneous pressures when the engine is fired or when motored. The motoring pressure is obtained by simulating the cycle while neglecting chemical reactions.

### Calibration of the Heat Transfer Coefficient

The heat transfer coefficient  $h$  is multiplied with a scaling factor  $\alpha$ , which is specific to the engine geometry. Hence, experimental data is required to calibrate the heat transfer model. To this end, the scaling factor was varied to fit the simulated pressure curve to a pressure curve from the literature [15]. In the publication, the sensitivity of HCCI engines to fuel compositions regarding autoignition characteristics under fuel-lean



**Fig. 1.** Comparison between the simulation with the fitted scaling factor and experiment [15] showing the pressure as a function of time and crank-angle. Simulation parameters:  $\phi = 0.3$ ,  $N = 1000$  rpm,  $T_0 = 442$  K,  $P_0 = 1.0$  atm.

conditions was investigated. Although their experiments were not carried out under partial oxidation (fuel-rich) conditions, they nevertheless provide useful data and enough information to allow simulations to be conducted for a calibration. The engine used in the experiment was a Volvo TD100 diesel truck engine, modified to run on a single cylinder. It has been used in numerous studies on HCCI engines, e.g. [15–17].

The relevant specifications of the simulated engine are given in Table 1. The air used in all simulations was composed as follows: 20.95 % oxygen, 78.12 % nitrogen and 0.93 % argon (simplified dry air, SDA). The composition of the natural gas was: 99 % methane, 1 % propane. The temperature of the cylinder walls was assumed to be 400 K, while the cylinder head and the piston were assumed to be 550 K.

The result of the fit is shown in Fig. 1. As can be seen, the model reproduces the experimentally obtained pressure curve reasonably well. However, there exists a pressure overshoot, which can be attributed to the single-zone model that does not include boundary layer effects [18]. Since the temperature of the boundary layer is significantly lower than in the bulk gas, the model over predicts the burn rate, also leading to an overestimated pressure rise.

**Table 1**  
Specifications of the simulated Volvo TD100 engine.

Bore	120.65 mm
Stroke	140 mm
Connecting Rod	260 mm
Displacement Volume	1.6 L
Compression Ratio	19.8

### Optimization Procedure

In a piston engine, the initial conditions can be varied to optimize the product composition at the end of the expansion stroke, i.e. maximize the conversion, the selectivity or the yield of certain chemical species. In particular, the inlet temperature  $T_0$ , the inlet pressure  $P_0$  and the equivalence ratio  $\phi$  (chemical composition) of the mixture can be controlled to maximize the hydrogen yield.

Hydrogen, being the more valuable product of the syngas mixture, is chosen as the objective function to be maximized, which is representative for syngas. Since CO is produced as a byproduct, the results are not expected to differ much if we choose to maximize CO instead of  $H_2$ . The objective function is the hydrogen yield at the end of the expansion stroke ( $t_f$ ) and the optimization problem can be formulated as follows:

$$\max Y_{H_2}(T_0, P_0, \phi) = \frac{2n_{H_2}(t_f)}{4n_{CH_4}(t_0) + 8n_{C_3H_8}(t_0)},$$

subject to the chemical dynamics

$$\frac{dn_i}{dt} = v \cdot \dot{\omega}_i,$$

the constraints

$$P(t) \leq 250 \text{ bar}, \quad \text{peak pressure}$$

$$t_{TC} \leq t_{\text{ignition}}, \quad \text{ignition timing}$$

and the bound constraints

$$300 \text{ K} \leq T_0 \leq T_{\text{max}},$$

$$1.0 \text{ bar} \leq P_0 \leq 5.0 \text{ bar},$$

$$0.1 \leq \phi \leq 5.0.$$

We define the time of ignition as the time where the temperature increase ( $dT/dt$ ) is highest.

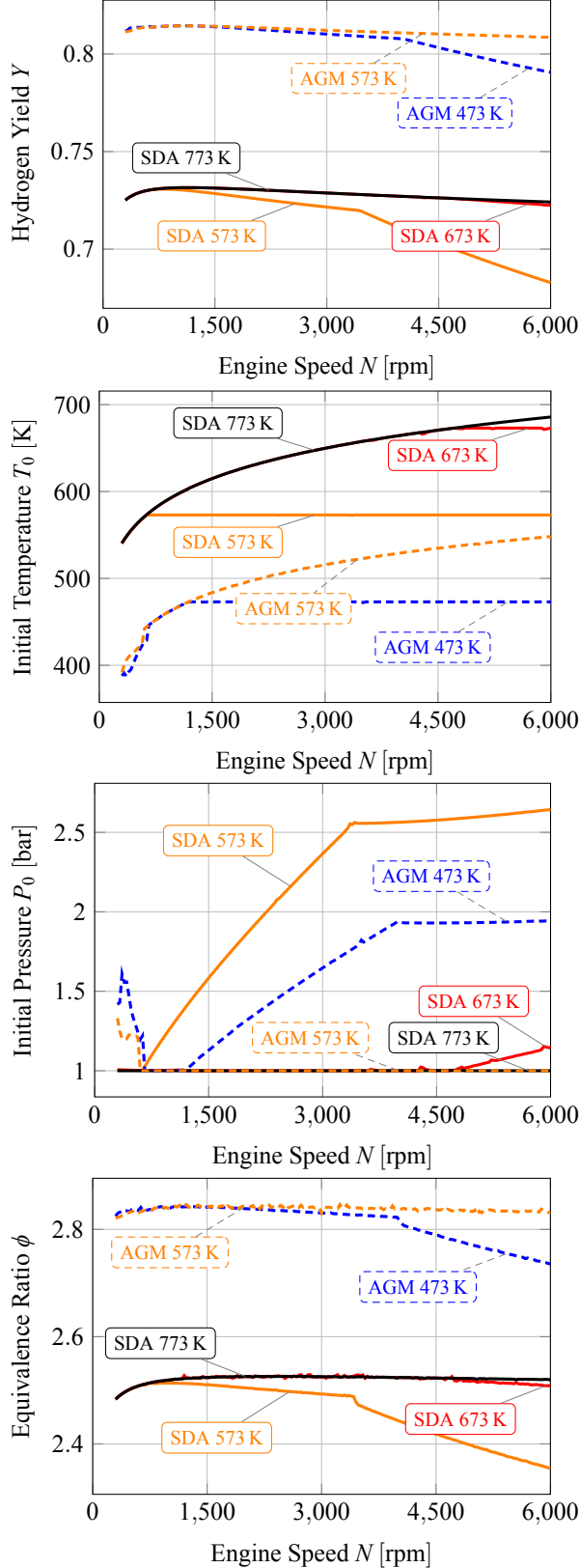
Powell’s COBYLA optimization algorithm [8], as implemented in the NLOpt library [9], was used to vary the optimization parameters  $T_0$ ,  $P_0$  and  $\phi$  and obtain an optimal hydrogen yield for a given engine speed. From these results, maximum syngas yield profiles and the associated operating parameter profiles as functions of engine speed were obtained. The engine speed was increased in steps of 30 rpm from 300 rpm up to 6000 rpm.

Additionally, suitable constraints were defined: the pressure must not exceed 250 bar and the ignition must occur after or at top center (TC). Furthermore, lower and upper bounds for each of the optimization variables were set. The profiles were calculated for a set of different upper temperature bounds  $T_{\text{max}}$ , varying from 473 K to 773 K (200 °C to 500 °C).

The COBYLA (constrained optimization by linear approximation) algorithm [8,9,19] is a local derivative-free optimization algorithm. It was chosen because it is easy to implement (in conjunction with the NLOpt library) and because it supports nonlinear constraints. Additionally, it performed favorably compared to other derivative-free algorithms such as the Nelder-Mead Simplex [20] or the Subplex algorithm [9,21]. The latter two algorithms were also used in preliminary test runs. However, it became apparent that nonlinear constraints were necessary to make the optimization problem meaningful. Hence no results obtained by these algorithms are included in this report. For example, it is necessary to constrain the temperature either directly via bounds or indirectly via the ignition timing constraint because the optimizer would otherwise strive towards unrealistically high temperatures. This is because thermodynamics dictates that high temperatures will favor the partial oxidation products  $H_2$  and CO instead of the fully oxidized products  $H_2O$  and  $CO_2$ .

### Optimization Results

Altogether, five optimal profiles as functions of engine speed were calculated and are shown in Fig. 2. The optimal hydrogen yield as a function of engine speed is shown in the top diagram and the three lower diagrams show the corresponding optimization variables  $T_0$ ,  $P_0$  and  $\phi$  that are required to obtain the objective function value. The line types indicate the oxidizer composition: solid for simplified air and dashed for an argon-enriched gas mixture (see below). The colors of the lines indicate the value of  $T_{\text{max}}$ .



**Fig. 2.** Optimization results showing optimal hydrogen yield (objective function), and corresponding optimization variables: initial temperature, initial pressure, equivalence ratio, each as a function of engine speed. The line types indicate the oxidizer composition: solid for simplified dry air (SDA) and dashed for the argon-enriched gas mixture (AGM). The colors of the lines indicate the value of  $T_{max}$ .

The results show that high temperatures favor high syngas yields. One possibility to attain higher temperatures is to decrease the mean heat capacity of the gas mixture. This can be accomplished by increasing the amount of argon in the reacting mixture. Since argon is a monoatomic gas, its heat capacity lies well below that of nitrogen. Thus, in addition to the three profiles with simplified dry air, two further optimization runs were calculated using an argon enriched gas mixture (AGM) as the oxidizing component: 20.95 %  $O_2$ , 70.95 % Ar. This composition corresponds to air where the nitrogen has been substituted by argon. This lead to higher syngas yields over a wide range of engine speeds. Furthermore, the use of AGM lead to an overall more stable engine operation at lower intake temperatures and higher speeds. However, it was much more difficult to satisfy the ignition timing constraint at low engine speeds.

While long residence times, i.e. low engine speeds, favor the hydrogen yield, they also make it difficult to time the ignition properly. In other words, low engine speeds can only be tolerated up to a certain point. If the engine is to be run even slower, then the ignition timing constraint forces the optimizer to choose conditions that are not favorable for high syngas yields. It is also particularly difficult to find a solution at these low speeds.

The temperature has a much higher impact than the initial pressure, which is consistent with the findings in [5]. This can be seen by the curves where the initial temperature is limited by the ignition timing constraint and not by the max temperature bound, e.g. using SDA with  $T_{max} = 573$  K (solid red curves). Only when a further increase in temperature is restricted by the bound constraints the initial pressure is increased. At low rpms, the ignition timing constraint restricts the temperature from being increased to the upper limit set by the bound constraint  $T_{max}$ . Only at engine speeds above 600 rpms does the bound constraint limit a further increase of initial temperature with engine speed. This can be seen well in the initial temperature profile, where the upper temperature bound constraint takes effect at 573 K. From this point onwards, the pressure is continuously increased with increasing engine speed. The sudden change in slope after around 3,500 rpm can be attributed to the peak pressure constraint, which implicitly limits the initial pressure.

Since the optimization algorithm yields locally optimal solutions, the optimization problem was solved multiple times with different initial guesses. Although different solutions were obtained in some cases, the hydrogen yield was very similar. In other words, many combinations are possible to obtain the same or a very similar objective function value. For example at 330 rpm, a hydrogen yield of 73 % can be achieved with an initial temperature of 540 K, an initial pressure of 1.47 bar and an equivalence ratio of 2.51. The same yield is possible with 573 K, 1.0 bar and an equivalence ratio of 2.51. This makes it difficult to pinpoint an exact optimal configuration. Hence, it is rather more appropriate to speak of regions. These circumstances pose advantages from a practical point of view, since it is

possible to choose from a multitude of combinations to achieve the same hydrogen yield. These results can therefore serve as a basis for a further optimization stage, when more technical constraints are known that narrow down the number of combinations further. In this study, the constraints were left intentionally wide as to make an evaluation regarding feasibility possible. Table 2 shows a combination of operating conditions to achieve a maximum hydrogen yield. However, different combinations are also possible which lead to the same result.

**Table 2**

Exemplary operating conditions to achieve maximum hydrogen yield. Different combinations are also possible.

	H <sub>2</sub> Yield	$\phi$	$N$ [rpm]	$T_0$ [K]	$P_0$ [bar]
SDA	73.1 %	2.52	1140	601	1.00
AGM	81.5 %	2.85	750	410	1.76

### Conclusions

In the present study, numerical optimization was applied to optimize the hydrogen yield in an HCCI piston engine fueled by natural gas. The optimization variables were the initial conditions of the process, i.e. the inlet temperature  $T_0$ , the inlet pressure  $P_0$  and the equivalence ratio  $\phi$ . We draw the following conclusions from the results of this study:

- The hydrogen yield does not vary much with increasing engine speed. This is beneficial because the production rate of hydrogen is linearly dependent on the engine speed. In other words, more hydrogen can be produced in the same amount of time without negatively affecting the conversion too much.
- The initial temperature has a much higher impact on the hydrogen yield than the initial pressure. However, increasing the pressure can also compensate for lower temperatures.
- A number of different combinations of the optimization parameters ( $T_0$ ,  $P_0$ ,  $\phi$ ) lead to a very similar objective function value. This allows a more flexible choice of operating conditions, e.g. those which are technically easier to achieve.
- The use of argon enriched air (AGM) makes it possible to run the engine at much lower initial temperatures. Further, the obtained hydrogen yield is almost 10% higher.

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