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Full Length Article

Autoignition studies of Liquefied Natural Gas (LNG) in a shock tube and a rapid compression machine

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ABSTRACT

Liquefied Natural Gas (LNG) has become an increasingly important world energy resource and is a part of the European Union clean fuel strategy launched in 2013. Therefore, there are currently several ongoing measurement strategies considering quality specification of LNG. In this context, for application in gas engines, it is essential to understand the combustion behavior of these natural gas mixtures. The methane number (MN) which represents a scale for the knocking propensity, is one of the main indicators for this combustion behavior. In this study, we investigated the influence of the LNG composition on the ignition delay time and thus the knocking behavior of prototypical LNG Mixtures. Several LNG typical mixtures containing CH_4/C_2H_6/C_3H_8/*n*-C_4H_{10}/*i*-C_4H_{10}/*n*-C_5H_{12}/*i*-C_5H_{12}/N_2 were studied in the temperature range 850–1450 K, with pressures of 20 and 40 bar and at equivalence ratios of 0.4 and 1.2. The use of a shock tube and a rapid compression machine facility allowed us to study the ignition behavior over a wide range of operating conditions relevant to gas engines. We report a detailed investigation of LNG autoignition with respect to temperature, pressure and equivalence ratio thereby providing crucial validation data for chemical kinetic models for real applications.

1. Introduction

Natural gas, as compressed natural gas (CNG) and Liquefied Natural Gas (LNG), is the fastest-growing fuel in the transportation sector, with a predicted average annual growth rate of 11.9% from 2011 to 2040. It is expected to emerge as a key transportation fuel during the next five years and will do more to slow down the oil demand growth than electric cars and biofuels combined [1,2].

LNG is generated at the extraction point of natural gas by cooling the gas to −162 °C. The liquid state generates several advantages for the use as transportation fuel over CNG. The energy density is much higher and it can be stored without significant overpressure thus not requiring heavy pressure tanks [3].

The composition of LNG, and consequently its energy content and other physical properties, varies from source to source. The composition mainly contains methane (79–99%) with a few percentages ethane and even lesser propane, *n*-butane, *iso*-butane and nitrogen as well as trace amounts of *n*-pentane and *iso*-pentane. The use of LNG as a fuel in an internal combustion engine can knocking and an increase in emissions and a decrease in the engine efficiency. Knocking is defined as abnormal combustion in an spark ignition (SI) engine, which occurs when a certain part of the fuel/air mixture auto-ignites and combusts before the arrival of the turbulent spark ignited flame. This phenomenon produces a rapid pressure rise and extremely high localized temperatures. This combination of high pressure and high temperature can cause erosion of the engine piston and is highly undesirable.

One of the primary factors influencing knock propensity of an engine is the fuel composition, which determines the rates of reactions and the heat release during the combustion process. Thus, a detailed analysis of the mixtures of LNG oxidation under engine relevant conditions, i.e., at high pressures and at intermediate temperatures is a key requirement for the characterization of LNG as transportation fuel.

Ignition of single component fuels like methane, ethane, propane, *n*-butane and *n*-pentane have been extensively investigated [4–8]. Whereas, ignition studies of multi-components fuels are scarce.

Healy et al. [9] studied the oxidation of methane/ethane/propane mixtures for blends containing 90/6.6/3.3, 70/15/15 and 70/20/10 percent by volume of each fuel, respectively, in air over the temperature range of 770–1580 K, at compressed gas pressures of approximately 1, 10, 20, 30, 40 and 50 atm, and at equivalence ratios of 0.5,

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1.0 and 2.0 using both, a high-pressure shock tube and a rapid compression machine. The experimental data were simulated using a detailed kinetic mechanism, which is based on the hierarchical nature of hydrocarbon combustion mechanism containing H_2/O_2, CO/CH_4 and larger hydrocarbon sub-mechanisms. Overall the mechanism was able to capture the experimental trends.

The same group [10] extended their study and investigated the oxidation of $CH_4/C_2H_6/C_3H_8/n\text{-}C_4H_{10}/n\text{-}C_5H_{12}$ mixtures in air over the temperature range of 630–1550 K, in the pressure range of 8–30 bar, and at equivalence ratios of 0.5, 1.0, 2.0 in both, a high-pressure shock-tube and in a rapid compression machine. The experimental data have been validated with a detailed chemical kinetic mechanism containing a comprehensive representation of low-temperature chemistry for fuels up to and including n-pentane. Overall, they observed good agreement between the model and experiments. However, a further improvement in the kinetic model is required to account for the discrepancies at high pressures and low temperatures.

One of the showstoppers in the roll-out of LNG as fuel is the lack of a commonly agreed methane number. The methane number is the LNG counterpart of the octane number for gasoline. However, no harmonized method to determine the methane number currently exists. A potentially suitable correlation between the methane number (MN) and the LNG composition is from MWM [11], which is an extension of the well-known AVL method [12]. The latter method is widely used, but has the disadvantage that it covers only hydrocarbons up to C_4 (butane). Gersen et al. [13] described a method 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. Using only a single, experimental non-knocking pressure trace, the method computes the effects of changes in autoignition chemistry, combustion phasing and thermophysical properties when varying the fuel, based only on the properties of the fuel–air mixture, using fuel-specific, detailed autoignition chemistry and burning velocity calculations.

To assess the knocking propensity of such fuels, the ignition delay time (IDT) of these fuels at typical operating conditions can be determined. The IDT is a measure for how easily a spontaneous ignition will occur. It is the time from when the fuel air mixture reaches the temperature and pressure which are sufficient to ignite it to when an actual macroscopic combustion can be observed.

The current work is designed to investigate the oxidation kinetics of mixtures containing $CH_4/C_2H_6/C_3H_8/n\text{-}C_4H_{10}/i\text{-}C_4H_{10}/n\text{-}C_5H_{12}/i\text{-}C_5H_{12}/N_2$. We have included data that was recorded in both a high-pressure shock tube and in a rapid compression machine for two equivalence ratios 0.4 and 1.2 at pressures of 20 and 40 bar in the temperature range of 850–1450 K. Chemical kinetic modeling was considered to get deeper insights into the relevant ignition chemistry.

2. Experiments

2.1. KAUST shock tube facility

Shock tube experiments were performed at the King Abdullah University of Science and Technology (KAUST), Saudi Arabia with the high-pressure shock tube (HPST). The details of this apparatus can be found in Ref. [14] and therefore only a brief overview is given here.

The HPST is constructed from stainless steel with an inner diameter of 10 cm. The driven section is 6.6 m long, and the driver has a maximum length of 6.6 m. The mid-section of the shock tube houses two pre-scored aluminium diaphragms in a double-diaphragm arrangement which allows for better control of the post reflected shock conditions compared to a single-diaphragm arrangement. The inner surface of the shock tube is electropolished to reduce the effects of boundary layers. The incident shock wave was monitored with five equally separated PCB 113B26 pressure transducers placed axially along the last 3.7 m from the driven section end-wall. The reflected-shock-test properties, i.e., P_5 and T_5, were calculated using one-dimensional normal shock relations. The uncertainties in the measured pressures and temperatures are less than 1%.

Table 1
Compositions (% volume) of the tested Liquid Natural Gas Mixtures in the shock tube as well as in the rapid compression machine.

Components	Mix-1	Mix-2
CH_4	87.89	78.80
C_2H_6	7.27	14.0
C_3H_8	2.92	3.40
$n\text{-}C_4H_{10}$	0.71	0.90
$i\text{-}C_4H_{10}$	0.65	1.10
$n\text{-}C_5H_{12}$	0.10	0.15
$i\text{-}C_5H_{12}$	0.11	0.15
N_2	0.35	1.5

A molar ratio of 3.76:1 of N_2:O_2 was used to prepare the typical LNG mixtures containing $CH_4/C_2H_6/C_3H_8/n\text{-}C_4H_{10}/i\text{-}C_4H_{10}/n\text{-}C_5H_{12}/i\text{-}C_5H_{12}/N_2$ in a magnetically-stirred mixing tank. Table 1 lists the compositions (% volume) of the two tested mixtures. The covered temperature range for these measurements was from 1100–1450 K. The shock tube and mixing tank were heated using heating jackets to avoid fuel condensation during the experiments.

The ignition onset was identified by pressure spike and by the maximum CH^* chemiluminescence detected at 430 nm using a Thorlabs PDA36A detector behind a narrow bandpass filter at the side and end wall of the shock tube. Fig. S1 depicts a schematic of the shock tube's detection system and typical recorded pressure and CH^* emission signal are represented in Fig. 1. The ignition delay time was determined from the signals by extracting the time difference between the passage of the reflected shock wave (time zero) to the steepest rate of change in the CH^* emission signal. The overall uncertainty in the shock tube ignition delay measurements was estimated to be ± 20%.

2.2. PTB rapid compression machine facility

The Rapid Compression Machine (RCM) experiments were performed at Physikalisch-Technische Bundesanstalt (PTB), Germany. The design is similar to the one designed by Mittal et al. [15]. It is a single piston rapid compression machine, which is pneumatically driven and hydraulically braked. A schematic can be found in Fig. S2 in the Supplementary Material. The RCM consists of three chambers, namely reactor, hydraulic and pneumatic chamber. The reactor chamber has an internal diameter of 50 mm and consists of 6 ports for pressure sensors,

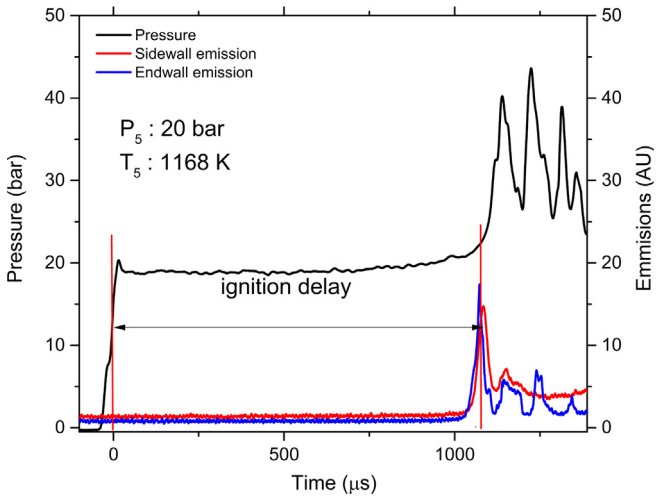

Fig. 1. Representative pressure and CH^* emission time profile for shock tube investigations.