



High-pressure shock-tube investigation of the impact of 3-pentanone on the ignition properties of primary reference fuels

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Abstract

Ignition-delay times for pure 3-pentanone, 3-pentanone/iso-octane (10/90% by volume) and 3-pentanone/*n*-Heptane mixtures (10/90% by volume) have been determined in a high-pressure shock tube under engine-relevant conditions ($p_5 = 10, 20, \text{ and } 40 \text{ bar}$) for equivalence ratios $\phi = 0.5$ and 1.0 and over a wide temperature range $690 \text{ K} < T_5 < 1270 \text{ K}$. The results were compared to ignition delay times of primary reference fuels under identical conditions. A detailed kinetics model is proposed for the ignition of all fuel mixtures. The model predicts well the ignition delay times for pure 3-pentanone for a wide range of pressure and temperature and equivalence ratios in argon dilution as well as in air. Ignition delay times for 3-pentanone-doped mixtures, especially in the low-temperature range are overpredicted by approx. a factor of 0.5 (at 800 K, 40 bar, $\phi = 1.0$) by the calculation but the model still reproduces the overall trend of the experimental data. For lean conditions, 10% 3-pentanone reduces the reactivity of *n*-Heptane below 1000 K while for stoichiometric conditions it does not alter the ignition delay by more than 11% at 850 K and 20 bar. In iso-octane the effect is inverse, leading to acceleration of the main ignition. Based on the model, the influence of 3-pentanone on the main heat release in a *n*-Heptane-fueled HCCI engine cycle is simulated.

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1. Introduction

Innovative concepts for internal combustion engines require a detailed knowledge about the gas mixture chemistry prior to ignition. Fluorescent tracers and fuel compounds are often used for fuel-concentration visualization based on laser-induced fluorescence (LIF) [1]. While the

temperature and pressure dependence of signals from these tracers have been studied in detail, studies about the oxidation chemistry of tracer-fuel mixtures are not well known and are just appearing.

Several species have been evaluated as LIF tracers. 3-pentanone (in the following, abbreviated as 3-P) has been identified as one “standard tracer” because it matches the boiling point and diffusion characteristics of the base fuel while having suitable optical characteristics. Its low sensitivity to collisional quenching by molecular oxygen is advantageous for quantitative signal

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interpretation. The photophysics of 3-P has been studied to derive quantitative data from fluorescence signals under varying ambient conditions. Modica et al. [2] investigated its signal dependence on temperature, pressure, and ambient gas composition. Neij et al. [3] used 3-P to map the fuel distribution at ignition in a spark-ignited (SI) engine using LIF. Han and Steeper [4] presented a method based on in-cylinder LIF measurements to identify the equivalence ratio in the presence of multi-component fuels. The technique was applied on different ketone tracers. Based on the T -dependent optical properties, Einecke et al. [5] demonstrated a two-line 3-P-LIF temperature-imaging technique for the unburned gas region of an SI engine. Rothamer et al. [6] used dual-wavelength excitation of 3-P in the ultraviolet to simultaneously measure exhaust gas residual mole fraction and temperature in an homogeneous-charge compression-ignition (HCCI) engine where both fuel and air were seeded with the tracer. A comprehensive review highlighting the different substances for the application of tracer LIF is given in [7].

Under high-temperature conditions, however, tracers exhibit limited stability and partially decompose together with the base fuel in the pre-ignition phase. Thus, the addition of the tracers to fuels for LIF experiments would have two important implications. On one hand, the measured fluorescence signals could be misinterpreted in case of the decomposition of the tracer, and, on the other hand, the tracer itself, depending on the conditions, can influence the ignition and combustion characteristics of the base fuel. Few papers addressed the interaction of fluorescence tracers and fuels. A theoretical study by Westbrook and Sick investigated the stability of biacetyl relative to reference fuels (primary reference fuels, PRF, i.e., mixtures from n -Heptane and iso-octane) based on kinetics models, however, without experimental validation [8]. Recently, we performed shock-tube measurements of ignition delay times to assess the influence of toluene (that is also frequently used as a fluorescent tracer) onto the ignition behavior of iso-octane and n -Heptane and to further validate the existing TRF (toluene reference fuel) mechanism [9]. Furthermore, numerical simulations were employed to study the decomposition of toluene under in-cylinder conditions and its significance as a representative for local fuel concentration and temperature or as an indicator for the time and the location of the onset of the heat release.

Fitzgerald et al. [10] showed in a direct injection HCCI engine, that for n -Heptane the chemical effects of tracer (e.g. 3-P and acetone) addition are more pronounced because of the effect of the low-temperature heat release. It was also shown that 3-P advances iso-octane combustion while retarding the ignition of n -Heptane. Serinyel et al. [11] studied the oxidation of 3-P/

O₂ mixtures in Ar at 1250–1850 K and atmospheric pressure behind reflected shock waves. Additionally, laminar flame speeds were measured in a spherical bomb and a kinetics mechanism for 3-P ignition was developed. The influence of the equivalence ratio was investigated and a correlation was derived. Unfortunately, this correlation does not account for the effect of pressure. Therefore, an extrapolation to higher pressures was not feasible. In addition, because Serinyel et al. [11] focused on the high-temperature range only, it was not possible to directly compare their results with those of the present work. Recently, Davidson et al. [12] investigated ignition delay times for a series of oxygenate species such as acetone, n -butanal, methyl butanoate, and 3-P, mainly motivated from their potential occurrence in bio-derived fuels. Again, the measurements covered only the high-temperature (>1173 K) and low-pressure ranges. These surveys identified the need for additional experimental data to cover the entire range relevant to IC-engine combustion and indicate the additional importance of oxygenates and their interaction with flame chemistry beyond the use of selected species as fluorescent tracers.

In this work, for the 3-P/PRF system, we extend the range of experimental conditions that has been published in [13] to lower temperatures and higher pressures to cover the conditions under which tracers are commonly employed and where their influence is supposed to be more significant. The experimental findings were compared with a blended model combining PRF and 3-P chemistry.

2. Experimental

The shock-tube facility used for gathering the ignition delay time data is described in detail in [14,15]. The shock tube has a constant inner diameter of 90 mm. The driver section is 6.4 m long and the driven section has a length of 6.1 m. The maximum test time is extended up to 15 ms by driver-gas tailoring. All experiments were conducted behind reflected shock waves in the 690–1270 K temperature range at pressures of 10, 20, and 40 bar. Measured ignition delay times of mixtures of the base fuels n -Heptane and iso-octane with 10% (by volume) 3-P were compared to those of the corresponding base fuels at two equivalence ratios $\phi = 0.5$ and 1.0. The concentration of 10% is commonly used for engine condition [16], 5% is doable in some cases [17].

The mixtures were prepared manometrically and settled for at least one hour to ensure homogeneous mixing. The influence of the mixing time on the ignition delay time for the facility used here has been studied before [18]. The temperature T_5 and pressure p_5 behind the reflected shock waves

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