



Auto-ignition of biomass synthesis gas in shock tube at elevated temperature and pressure

Linqi Ouyang · Hua Li · Shuzhou Sun ·
Xiaole Wang · Xingcai Lu

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Abstract Ignition delay times of multi-component biomass synthesis gas (bio-syngas) diluted in argon were measured in a shock tube at elevated pressure (5, 10 and 15 bar, 1 bar = 10^5 Pa), wide temperature ranges (1,100–1,700 K) and various equivalence ratios (0.5, 1.0, 2.0). Additionally, the effects of the variations of main constituents ($H_2:CO = 0.125\text{--}8$) on ignition delays were investigated. The experimental results indicated that the ignition delay decreases as the pressure increases above certain temperature (around 1,200 K) and vice versa. The ignition delays were also found to rise as CO concentration increases, which is in good agreement with the literature. In addition, the ignition delays of bio-syngas were found increasing as the equivalence ratio rises. This behavior was primarily discussed in present work. Experimental results were also compared with numerical predictions of multiple chemical kinetic mechanisms and Li's mechanism was found having the best accuracy. The logarithmic ignition delays were found nonlinearly decrease with the H_2 concentration under various conditions, and the effects of temperature, equivalence ratio and H_2 concentration on the ignition delays are all remarkable. However, the effect of pressure is relatively smaller under current conditions. Sensitivity analysis and reaction pathway analysis of methane showed that R1 ($H + O_2 = O + OH$) is the most sensitive reaction promoting ignition and R13 ($H + O_2 (+M) = HO_2 (+M)$), R53

($CH_3 + H (+M) = CH_4 (+M)$), R54 ($CH_4 + H = CH_3 + H_2$) as well as R56 ($CH_4 + OH = CH_3 + H_2O$) are key reactions prohibiting ignition under current experimental conditions. Among them, R53 ($CH_3 + H (+M) = CH_4 (+M)$), R54 ($CH_4 + H = CH_3 + H_2$) have the largest positive sensitivities and the high contribution rate in rich mixture. The rate of production (ROP) of OH of R1 showed that OH ROP of R1 decreases sharply as the mixture turns rich. Therefore, the ignition delays become longer as the equivalence ratio increases.

Keywords Shock tube · Biomass synthesis gas · Ignition delay time · Sensitivity analysis · Reaction pathway analysis

1 Introduction

Biomass synthesis gas with carbon-neutral property has been paid extensive attention because of the severe pollution in some countries at present. Biomass synthesis gas can be utilized in various energy power devices—it not only can be used in biomass gasification power generation and combined cooling, heating and power (CCHP), but also used as clean alternative fuels in internal combustion engines, fuel cells and boilers [1, 2]. It can be promising that biomass synthesis gas will occupy a critical position in the future energy fields. Generally, the feedstocks which can be utilized to produce synthesis gas include natural gas, coal and biomass (such as feces of animals, straw and other organic wastes) [3, 4]. In China, due to the emission caused by the burning of crop straw [5], it is absolutely a promising method to make use of straw by turning them into synthesis gas. The synthesis gas derived from biomass is called biomass-derived syngas or bio-syngas. Recently, the bio-syngas has aroused great

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L. Ouyang · H. Li · S. Sun · X. Wang · X. Lu (✉)
Laboratory for Power Machinery and Engineering of Ministry of Education, Shanghai Jiao Tong University, Shanghai 200240, China
e-mail: lyuxc@sjtu.edu.cn

interest among researchers because of the diversity and availability of feedstocks [6–12].

Practically, the main components of bio-syngas are CO and H₂. Other components (including methane, nitrogen, carbon dioxide, water [9, 12]) differ a lot due to the diversity of biomass feedstocks and production procedures of bio-syngas. The combustion characteristics will alter since the components in bio-syngas vary. Therefore, the power output and emission of internal combustion engines and gas turbines will be affected, which could be adverse to the design of engines and control strategies [13]. No matter if bio-syngas is used in internal combustion engines or in gas turbines, the ignition properties and flame properties of bio-syngas could have great effects on the efficiency and emission of the whole combustion system. Mittal et al. [14] studied the ignition delay times of 80 % CO + 20 % H₂ mixtures in rapid compression machine (RCM) at temperature of 950–1,050 K and pressure of 15, 30 and 50 bar. Gersen et al. [15] measured the ignition delay times of stoichiometric and lean H₂, H₂ + CO, CH₄, CH₄ + CO, CH₄ + H₂ and CH₄ + CO + H₂ mixtures at pressures ranging from 20 to 80 bar and in the temperature range 900–1,100 K in a RCM. The effects of the content of CO (<50 %) and CH₄ on the ignition delays of hydrogen were investigated. Walton et al. [16] also investigated the ignition of simulated syngas mixtures in a rapid compression facility at spanned pressures ranging from 7.1 to 26.4 atm, temperatures from 855 to 1,051 K, equivalence ratios from 0.1 to 1.0, oxygen content from 15 % to 20 % and H₂:CO from 0.25 to 4.0. Mansfield and Wooldridge [17] investigated the effects of chemical impurities (CH₄, TMS) on the combustion of syngas in a rapid compression facility. Two-step ignition behavior was found at higher pressures, and the ignition delays were affected differently by the addition of impurities. Thi et al. [18] measured the ignition delay times of syngas in a shock tube at temperature ranging from 757 to 1,280 K, equivalence ratios ranging from 0.3 to 1.0 and pressure of 2 and 10 bar. Sivaramakrishnan et al. [19] measured ignition delay times of dilute CO/H₂ mixtures in a high-pressure shock tube. They also analyzed variation of some stable species (CO, O₂, CO₂) using GC and GC/MS. Petersen et al. [20] studied the ignition delays of CO/H₂/CO₂ mixtures using shock tube and flow reactor. Results were compared with previous work of Peschke and Spadaccini [21] indicating that their variations of ignition delay times over time agreed well, but there are deviations between experimental results and predictions. Mathieu et al. [22, 23] studied the ignition delay times of syngas derived from coal and biomass in a shock tube. The effects of other components (CH₄, CO₂, H₂O, NH₃, H₂S) other than CO and H₂ on the ignition delays were investigated.

In terms of the combustion flame characteristics of syngas, Wang et al. [24] measured the transient front of

turbulent premixed flame of biomass syngas (CO/H₂/CO₂/air) by using PLIF. Prathap et al. [25] studied the effect of dilution with CO₂ on the laminar burning velocity and flame stability of syngas (50 % hydrogen + 50 % CO) in a constant-pressure spherical diffusion flame. The results showed that peak burning velocity shifts because of dilution with CO₂. CO₂ shows a stronger inhibiting effect on laminar burning velocity than that of nitrogen possibly due to the participation of CO₂ in the chemical reactions. As for the chemical effects of CO₂, Liu et al. [26] numerically studied the chemical effects of CO₂ replacement of N₂ in air on the burning velocity of CH₄/O₂/N₂/CO₂ and H₂/O₂/N₂/CO₂ mixtures. Bouvet et al. [27] particularly measured the spherical flame velocity of syngas at spanned H₂/CO ratios ranging from 5 %/95 % to 50 %/50 % and equivalence ratios ranging from 0.4 to 5.0. Natarajan et al. [28] investigated the burning velocity of H₂/CO/CO₂ syngas mixtures using conical Bunsen flame and one-dimensional stagnation flame at a wide range of fuel fractions (fraction of H₂ + CO spanning from 5 % to 95 %, fraction of CO₂ is less than 40 %). Dong et al. [29] experimentally measured the laminar flame velocity of H₂/CO mixtures (volume fraction of H₂ ranging from 0 to 100 %) in a Bunsen burner. Goswami et al. [30] measured the laminar flame speed of H₂/CO and H₂/N₂ mixtures in O₂/He. Krejci et al. [31] studied the laminar flame speed of the neat H₂ and CO/H₂ mixtures. Hydrogen, as a reactive component in syngas, was also experimentally and numerically studied with air in a closed combustion vessel [32]. Tinaut et al. [33] studied the combustion velocity of biomass producer gas (CO/CO₂/CH₄/H₂/N₂) with various gas concentrations in a constant-volume combustion bomb.

Multitudes of scholars have developed the chemical kinetics of CO/H₂ mixtures due to the importance of chemical kinetic characteristics of bio-syngas [34–37]. In addition, Wang et al. [38] developed a detailed chemical kinetic mechanism of mixtures containing CO, H₂ and other species. Dryer and Chaos [39] investigated the homogeneous gas-phase combustion kinetics of syngas at low temperatures and high pressures ($T < 1,000$ K, $10 < P < 30$ atm). Boivin et al. [40] developed a four-step reduced chemical kinetic model based on CO + H₂ bio-syngas, specifically for combustion in gas turbine. Petersen et al. [20] performed an investigation on the ignition of syngas at real conditions, and the predictions of previous chemical kinetics were found to be deviating sharply from results at real conditions. The major reason is that the main components in syngas are CO, H₂, CO₂ and other gases; however, the widely studied H₂/CO mechanisms lack investigation at high pressure and low temperature. Kérômènes et al. [41] measured the ignition delay times of H₂/CO/O₂/N₂/Ar mixture in shock tubes and two rapid compression machines at pressures ranging from 1 to 70 bar,

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