



A further experimental and modeling study of acetaldehyde combustion kinetics

Tao Tao^a, Shiqing Kang^a, Wenyu Sun^a, Jiaxing Wang^a, Handong Liao^a, Kai Moshhammer^{c,d}, Nils Hansen^c, Chung K. Law^{a,b}, Bin Yang^{a,*}

^a Center for Combustion Energy and Key Laboratory for Thermal Science and Power Engineering of MOE, Department of Energy and Power Engineering, Tsinghua University, Beijing 100084, PR China

^b Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, NJ 08544, USA

^c Combustion Research Facility, Sandia National Laboratories, Livermore, CA 94551, USA

^d Physikalisch-Technische Bundesanstalt, Department of Thermophysical Quantities, Bundesallee 100, 38116 Braunschweig, Germany

ARTICLE INFO

Article history:

Received 14 February 2018

Revised 5 June 2018

Accepted 6 June 2018

Keywords:

Acetaldehyde

Counterflow flame

Ignition delay times

Negative temperature coefficient

Kinetic model

ABSTRACT

Acetaldehyde is an important intermediate and a toxic emission in the combustion of fuels, especially for biofuels. To better understand its combustion characteristics, a detailed chemical kinetic model describing the oxidation of acetaldehyde has been developed and comprehensively validated against various types of literature data including laminar flame speeds, oxidation and pyrolysis in shock tubes, chemical structure of premixed flames, and low-temperature oxidation in jet-stirred reactors. To extend the validation range, the chemical structure of a counterflow flame fueled by acetaldehyde at 600 Torr has been measured using vacuum ultra-violet photoionization molecular-beam mass spectrometry. In addition, ignition delay times at 10 atm and 700–1100 K were measured in a rapid compression machine, and a negative temperature coefficient (NTC) behavior was observed. The present kinetic model well reproduces the results of various acetaldehyde combustion experiments covering wide ranges of temperatures (300–2300 K) and pressures (0.02–10 atm), and explains well the observed NTC behavior based on the competition between multiple oxidation pathways for the methyl radicals and their self-recombination forming ethane, a relatively stable species at temperatures below 1000 K.

© 2018 The Combustion Institute. Published by Elsevier Inc. All rights reserved.

1. Introduction

Acetaldehyde plays an important role in biofuel combustion as one of the most abundant toxic, oxygenated emissions [1,2]. For example, experimental studies on the structure of the premixed flames fueled by butanols show that acetaldehyde concentration could be as high as 6000 ppm [3]. A comprehensive chemical kinetic mechanism describing the oxidation and pyrolysis behavior of acetaldehyde is crucial to the construction of combustion kinetic models for practical fuels, especially for biofuels, and can also contribute to improved understanding and prediction of pollutant formation.

Several kinetic models have been proposed to describe the oxidation or pyrolysis of acetaldehyde. Specifically, the model of Kaiser et al. [4] described acetaldehyde oxidation in the negative temperature coefficient (NTC) regime between 550 and 900 K, while the model of Pelucchi et al. [5,6] well reproduced the

observed ignition and cool flame behaviors of mixtures of acetaldehyde and oxygen from 450 to 625 K [7]. Dagaut and co-workers [8] proposed a model to describe the oxidation in a jet-stirred reactor (JSR) and the ignition in a shock tube (ST) covering medium to high temperature ranges (900–2530 K). A high-temperature model developed by Leplat and Vandooren [9] was validated against their speciation measurements in low-pressure premixed flames. The AramcoMech 2.0 [10–14], developed as a core mechanism, has been tested against the ignition delay times of acetaldehyde measured by Yasunaga et al. [15] and the speciation results during the oxidation at 1100 K in a flow tube [16]. Based on theoretical calculations, Sivaramakrishnan et al. [17] built a model for the pyrolysis of acetaldehyde. In spite of such extensive and worthy efforts, it is nevertheless noted that they were all developed based on only one or two sets of experimental results, hence limiting the validation range for parameters. Mével et al. [18] published a widely-validated kinetic model, tested against the oxidation and pyrolysis results in STs [15,19,20], laminar flame speeds [21], oxidation in JSR [8] and speciation measurement in low-pressure premixed flames [22]. This model, however, focuses on the high-temperature pyrolysis and oxidation

* Corresponding author.

E-mail address: byang@tsinghua.edu.cn (B. Yang).

of acetaldehyde, containing no reactions related to the low-temperature oxidation. Recently, speciation of the acetaldehyde oxidation in a JSR was measured by Zhang et al. [23] between 460 and 900 K. A model for acetaldehyde oxidation was also proposed by incorporating theoretically calculated rate coefficients for the H-abstractions from CH_3CHO by CH_3O , CH_3O_2 and CH_3CO_3 radicals. In their model, a pathway of second O_2 -addition to CH_3CO radical was included to explain the high reactivity of acetaldehyde oxidation at low temperatures, although, this pathway was theoretically found by Maranzana et al. [24] to be negligible in the oxidation of CH_3CO radicals. A comprehensively validated kinetic model for acetaldehyde combustion that covers the combined low- and high-temperature chemistries is therefore needed.

In terms of experimental investigations, it is noted that over the past decades, the oxidation and pyrolysis of acetaldehyde have been experimentally investigated in STs [8,15,18–20,25,26], laminar premixed flames [9,21,22], JSRs, and flow reactors [16,27]. In 1981, Gray et al. [7] reported the oscillatory ignition and cool flames phenomenon in the acetaldehyde oxidation. Subsequently, Kaiser et al. [4] investigated the NTC behavior in the low-temperature oxidation of acetaldehyde, providing information about the oxidation process based on their measurements at two temperatures (553 and 713 K). Recently, Tao et al. [28] and Zhang et al. [23] separately reported the species concentration profiles measured in the low-temperature oxidation of acetaldehyde in JSRs, thus broadening the kinetic understanding of the NTC behavior of acetaldehyde. The NTC behavior of various fuels has been found [29–32] in the measured ignition delay times, but until now, such observation has not yet been reported for acetaldehyde. It is therefore meaningful to investigate the manifestation of the NTC phenomenon in the ignition delay times and explore the associated oxidation kinetics.

As for the high-temperature oxidation of acetaldehyde, detailed experimental information is also limited. Besides the measurements of the global characteristics, such as the laminar flame speeds [21] and the ignition delay times [8,15,18,25], Leplat and Vandooren [9] measured the mole fraction profiles of five intermediates in the laminar premixed flames of acetaldehyde. We recently extended this flame work by employing vacuum ultra-violet photoionization molecular-beam mass spectrometry (VUV-PI-MBMS) to determine the detailed chemical structure of two low-pressure laminar premixed flames [22], recognizing nevertheless that these low-pressure flame results are not likely to be sufficient to unambiguously examine the high-temperature sub-mechanism of acetaldehyde combustion. Detailed experimental results from different flame structures or under different pressures, for example as those in the counterflow flames, would facilitate the validation of a comprehensive model.

In the present study, a kinetic model for the oxidation of acetaldehyde was developed as a continuing effort of our previous work [28], in which a modified model based on AramcoMech 2.0 was proposed to describe and explain the low-temperature oxidation of acetaldehyde, especially the NTC behavior. The performance of the present kinetic model has been comprehensively examined with several sets of experimental data in the literature [8,15,21,22,26,28]. Furthermore, to extend the validation range, the ignition delay times at medium temperatures (700–1100 K) were measured in a rapid compression machine (RCM) and the species concentration profiles in a counterflow flame fueled by acetaldehyde were measured by VUV-PI-MBMS.

2. Experimental specifications

2.1. Rapid compression machine

The ignition delay times of acetaldehyde were measured in the RCM at Tsinghua University, covering the temperature range of

Table 1

Mixture compositions and experimental conditions.

	Equivalence ratio	Mole fractions (%)				Pressure bar	Temperature K
		CH_3CHO	O_2	Ar	N_2		
1	1.0	2.78	6.92	90.30	/	10	734–1086
2	–	2.78	/	90.25	6.97		

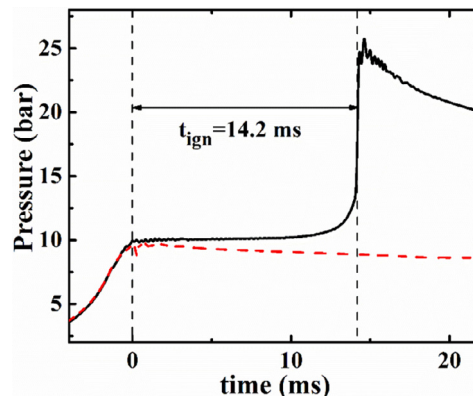


Fig. 1. Pressure trace history of reactive (black solid line) and nonreactive mixture (red dashed line) in the RCM at $P_{\text{eff}} = 10$ bar, $T_{\text{eff}} = 851$ K, $\Phi = 1.0$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

734–1086 K at 10 bar with the equivalence ratio of 1.0. The configuration of the RCM has been described in details by Di et al. [33]. The gaseous mixture was prepared in a stainless steel mixing tank, whose composition was determined according to the partial pressure of each mixture component. The partial pressure of acetaldehyde was controlled to be less than 50% of its vapor pressure to avoid potential condensation. Argon was selected as the dilution and the diluent ratio (Ar/O_2) was fixed at 13 to obtain a wide test temperature range. The mixture composition and experiment conditions are listed in Table 1. To calibrate the heat-loss effect after the compression, a nonreactive mixture with similar heat capacity was prepared by replacing O_2 with N_2 , as shown in Table 1. Since the partial pressure of each component cannot be controlled exactly in the preparation of each mixture, there is a slight difference in the concentrations of the two mixtures. Pressure traces were recorded for both mixtures under the same conditions.

Figure 1 displays a typical pressure trace profile using both mixtures at the same condition. Different from the situations for other fuels, the heat release rate of acetaldehyde oxidation before ignition is larger than the heat loss rate caused by heat conduction, due to the high activity of acetaldehyde oxidation especially at temperatures lower than 900 K. Consequently, after the end of compression (EOC), the pressure increases slowly, rather than decreasing as in the cases for common fuels. The instant of EOC was determined as the first peak pressure after compression in the recorded pressure history of the nonreactive mixture. The effective pressure ($P_{\text{eff}} = 10$ bar) was determined as the pressure at the EOC point instead of the integrated average of pressure from the first peak pressure to the minimum pressure after compression, as explained in Refs [33,34]. Based on the isentropic assumption, the effective temperatures ($T_{\text{eff}} = 734$ –1086 K) were calculated from the pressure by considering the corresponding compression ratios. The ignition delay time was defined as the time interval between EOC and the state of the maximum rate of the pressure rise. The method for the uncertainty evaluation has been described in Ref. [33]. The measured ignition delay times with their

Download English Version:

<https://daneshyari.com/en/article/6593380>

Download Persian Version:

<https://daneshyari.com/article/6593380>

[Daneshyari.com](https://daneshyari.com)