



New insights in the low-temperature oxidation of acetylene

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Abstract

This work presents new experimental data of C_2H_2 low-temperature oxidation for equivalence ratios $\Phi = 0.5$ – 3.0 in a newly designed jet-stirred reactor over a temperature range of 600–1100 K at atmospheric pressure with residence time corresponding from 1.94 to 1.06 s. Mole fraction profiles of 17 intermediates including aromatic compounds such as toluene, styrene and ethylbenzene were quantified. A detailed kinetic mechanism involving 295 species and 1830 reactions was established to predict the oxidation of C_2H_2 and formation of PAH. In developing the mechanism, particular attention was paid to reactions of the vinyl radical and to steps involved in the sequence $C_2H_2 \rightarrow iC_4H_5 \rightarrow$ fulvene $\rightarrow C_5H_5CH_2 \rightarrow C_6H_6$. In general, the peak concentrations of intermediates gradually increase and peak locations tend to shift toward high temperatures with Φ increasing. Flux analysis indicates that the addition of H and the reaction with O are the two major channels governing C_2H_2 consumption. At temperatures below 1000 K, benzene is mainly formed through the $C_2 + C_4$ channels: $C_2H_2 + iC_4H_5 \rightarrow$ fulvene $\rightarrow C_5H_5CH_2$ isomers $\rightarrow C_6H_6$. The $C_1 + C_5$ pathway: $CH_3 + C_5H_5 \rightarrow C_5H_5CH_3 \rightarrow$ (fulvene and $C_5H_5CH_2$ radicals) $\rightarrow C_6H_6$ tends to be the dominant route for benzene formation at temperatures above 1000 K. In addition to the present data, the model predicts well ignition delay times reported in literature.

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1. Introduction

The interest for studying the low-temperature oxidation of C_2H_2 has increased in recent years.

C_2H_2 is a significant intermediate during the combustion of many hydrocarbon fuels, particularly under fuel-rich conditions. It serves as a key precursor leading to the formation of polycyclic aromatic hydrocarbons (PAH) and soot [1], which are combustion-derived air pollutants known to be harmful to both the human health and environment. Consequently, experimental and

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modeling studies on C_2H_2 oxidation are important to improve our knowledge on the combustion of hydrocarbons, in particular formation of PAH and soot.

Previous studies of C_2H_2 were mainly focused on flame and pyrolysis experiments, providing a large body of data for validation of C_2H_2 models. From 1982, a number of laminar premixed flame studies of C_2H_2 at near sooting limit have been reported [2–11]. Among studies addressing C_2H_2 pyrolysis, several have been conducted in shock tubes [12–17]. Compared to the flame and pyrolysis investigations, oxidation studies of C_2H_2 in flow reactors are relatively scarce; only the work by Tan et al. [18] in a jet-stirred reactor (JSR) and Alzueta et al. [19] in a flow reactor are available. Tan et al. [18] studied C_2H_2 oxidation at 800–1100 K and 1 to 10 atm at fuel/air equivalence ratios $\Phi = 0.4$ to 1.1 and reported profiles of H_2 , CO, CO_2 , CH_4 , C_2H_2 , C_2H_4 and pC_3H_4 . A comprehensive model was proposed to reproduce the measured data. However, aromatic species were not detected in their experiments. Alzueta et al. [19] performed C_2H_2 oxidation experiments for $\Phi = 0.05$ to 1.43 at atmospheric pressure in 700–1400 K and measured C_2H_2 , CO, CO_2 and C_2H_4 . While the overall oxidation behavior was predicted fairly well, PAH and soot formation were not considered.

Low-temperature oxidation data of C_2H_2 are limited in the literature, especially under fuel-rich conditions. Experiments at higher values of Φ , together with a more comprehensive species characterization, will be helpful in improving the understanding of aromatics formation. This work aims to identify and quantify the intermediate and product composition of C_2H_2 low-temperature oxidation and to develop a comprehensive model for C_2H_2 oxidation under wide range of stoichiometry. Experiments on oxidation of C_2H_2 were conducted in a jet-stirred reactor (JSR) in the temperature range of 600–1100 K and for $\Phi = 0.5$ –3.0 with residence time from 1.94 to 1.06 s at atmospheric pressure. Mole fraction profiles of reactants, intermediates and products were measured through continuous sampling by gas chromatography (GC) analysis. Moreover, predictions with seven published models and the current updated mechanism were compared to the experimental data, as well as data from literature. The results extend the experimental database for C_2H_2 oxidation, particularly under fuel-rich conditions, and provide insight into the low temperature oxidation mechanism and formation of PAH.

2. Experimental

The experiments were performed in a spherical fused-silica JSR with 50 mm inner diameter at atmospheric pressure within 600–1100 K. In the experiments, 0.4% C_2H_2 and 0.33–2.0% O_2 diluted in

Table 1
Experimental conditions.

Φ	C_2H_2 (%)	O_2 (%)	Ar (%)	TFR ^a (sccm)
0.5	0.40	2.00	97.60	1000
1.0	0.40	1.00	98.60	1000
2.0	0.40	0.50	99.10	1000
3.0	0.40	0.33	99.27	1000

^a TFR refers to total flow rate.

Ar, corresponding to $\Phi = 0.5$ –3.0, were introduced into the reactor at a constant flow rate of 1000 standard cubic centimeter per minute (sccm). Compared to the previously reported JSR work by Tan et al. [18] (0.15% C_2H_2 , $\Phi = 0.4$ –1.2, 800–1100 K), a higher concentration of C_2H_2 , and wider ranges of temperature and equivalence ratios were used in this work. The experiments under very reducing conditions allowed detection of a number of products not previously reported for oxidation of C_2H_2 in the current temperature range. The apparatus, newly built in the Institute of Engineering Thermophysics, Chinese Academy of Sciences, is displayed in Fig. S1 in the Supplementary Material (SM) and the experimental conditions are summarized in Table 1 over the temperature range of 600–1100 K with corresponding residence time from 1.94 to 1.06 s. The reliability of the constructed apparatus was validated by reproducing the low temperature oxidation of C_2H_2 in JSR by Tan et al. [18] and acceptable results were obtained.

To argue that the perfectly stirred reaction condition can be obtained both in concentration and temperature, the following fluid mechanical calculation was performed. The homogeneity of the composition in the JSR relies on three criteria: the turbulent jets from the four nozzles, a good mixing of the composition by the four jets and the velocity of the outlet jets not exceeding the speed of sound (C_{sound}). The first and the third criteria fix the range of accessible residence times and the second criterion fixes the geometrical ratio between the internal diameters of the reactor and of the nozzles. To verify the criteria of the JSR used, argon was applied as the carrier gas. Verification was performed at 298 K and atmospheric pressure. The radius of the reactor, R , and the internal diameter of the nozzle, d , are taken equal to 25 and 0.3 mm, respectively. The upper limit for the residence time is given by equation: $\tau \leq \rho AR^3/230\eta d$ [20]. The specific weight and the dynamic viscosity of argon are equal to 1.61 kg/m³ and 2.26 $\times 10^{-5}$ Pa s, respectively. The value of the parameter A for air at 298 K is used ($A = 0.3$). The calculation leads to a maximum residence time of 4.9 s. The lower limit is given by equation $\tau \geq 4R^3/3d^2 C_{\text{sound}}$ [20]. C_{sound} for argon at 298 K equals to 321 m/s. The calculation leads to a minimum residence time of 0.7 s. The recycling rate is given by equation $Rr = \pi AR/2d$. Calculation leads to a recycling rate of 103 ($A = \pi/4$), which is larger than the minimum required value of 30 to

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