



Rich methane premixed laminar flames doped with light unsaturated hydrocarbons

I. Allene and propyne

H.A. Gueniche, P.A. Glaude, G. Dayma, R. Fournet, F. Battin-Leclerc *

Département de Chimie-Physique des Réactions, Nancy University, CNRS, ENSIC, 1 rue Grandville, BP 20451, 54001 Nancy Cedex, France

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Abstract

The structure of three laminar premixed rich flames has been investigated: a pure methane flame and two methane flames doped by allene and propyne, respectively. The gases of the three flames contain 20.9% (molar) of methane and 33.4% of oxygen, corresponding to an equivalence ratio of 1.25 for the pure methane flame. In both doped flames, 2.49% of C_3H_4 was added, corresponding to a ratio C_3H_4/CH_4 of 12% and an equivalence ratio of 1.55. The three flames have been stabilized on a burner at a pressure of 6.7 kPa using argon as dilutant, with a gas velocity at the burner of 36 cm/s at 333 K. The concentration profiles of stable species were measured by gas chromatography after sampling with a quartz microprobe. Quantified species included carbon monoxide and dioxide, methane, oxygen, hydrogen, ethane, ethylene, acetylene, propyne, allene, propene, propane, 1,2-butadiene, 1,3-butadiene, 1-butene, isobutene, 1-butyne, vinylacetylene, and benzene. The temperature was measured using a PtRh (6%)–PtRh (30%) thermocouple settled inside the enclosure and ranged from 700 K close to the burner up to 1850 K. In order to model these new results, some improvements have been made to a mechanism previously developed in our laboratory for the reactions of C_3 – C_4 unsaturated hydrocarbons. The main reaction pathways of consumption of allene and propyne and of formation of C_6 aromatic species have been derived from flow rate analyses.

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1. Introduction

Soots and polyaromatic hydrocarbons (PAH), which are present in the exhaust gas of diesel en-

gines, represent a large part of the urban pollution. Many efforts have then been focused on reducing the emissions of these compounds. The formation of soot precursors and PAH in combustion involves small unsaturated hydrocarbons, the chemistry of which is still very uncertain. Different reaction pathways have been proposed for the formation and the oxidation of the first aromatic compounds, involving the reactions of C_2 (acetylene), C_3 , or C_4 unsaturated species [1–5].

* Corresponding author. Fax: +33 3 83 37 81 20.

E-mail address:

frederique.battin-leclerc@ensic.inpl-nancy.fr

(F. Battin-Leclerc).

As the determinant role of propargyl radicals in forming benzene, the first aromatic ring, is now well accepted, it is important to better understand their reactions. With that purpose, the oxidation of allene (propadiene) and propyne has been already studied in several oxidation conditions: shock tubes [6–8], flow reactor [9,10], jet-stirred reactor [7,11] and premixed flames [10,11]. Previous work in flames includes studies of the influence of the addition of allene on $H_2/O_2/Ar$ [12], $C_2H_2/O_2/Ar$ [13], and $C_2H_4/O_2/Ar$ [14] mixtures.

The purpose of the present paper is to experimentally investigate the structures of two premixed laminar methane flames containing propadiene and propyne, respectively, and to compare them with that of a pure methane flame containing the same mole fractions of methane and oxygen. The use of a methane flame will allow us to have a reactive mixture rich in methyl radicals and more representative of combustion mixtures containing larger hydrocarbons than hydrogen or C_2 flames. These results have been used to improve the mechanism previously developed in our laboratory for the reactions of C_3 – C_4 unsaturated hydrocarbons (propyne, allene, 1-butyne, 2-butyne, 1,3-butadiene) [8,15].

2. Experimental procedure

The experiments were performed using an apparatus newly developed in our laboratory to study temperature and stable species profiles in a laminar premixed flat flame at low pressure. The body of the flat flame matrix burner, provided by McKenna Products, was made of stainless steel, with an outer diameter of 120 mm and a height of 60 mm (without gas/water connectors). This burner was built with a bronze disk (95% copper, 5% tin). The porous plate (60 mm diameter) for flame stabilization was water-cooled (water temperature: 333 K) with a cooling coil sintered into the plate. The burner could be operated with an annular co-flow of argon to favor the stabilization of the flame.

This horizontal burner was housed in a water-cooled vacuum chamber evacuated by two primary pumps and maintained at 6.7 kPa by a regulation valve. This chamber was equipped with four quartz windows for an optical access, a microprobe for samples taking and a thermocouple for temperature measurements. The burner could be vertically translated, while the housing and its equipments were kept fixed. A sighting telescope measured the position of the burner relative to the probe or the thermocouple with an accuracy of 0.01 mm. The flame was lighted on using an electrical discharge. Gas flow rates were regulated by RDM 280 Alphagaz and Bronkhorst (El-

Flow) mass flow regulators. The C_3 reactants (purity discussed in the text) and methane (99.95% pure) were supplied by Alphagaz–L'Air Liquide. Oxygen (99.5% pure) and argon (99.995% pure) were supplied by Messer.

Temperature profiles were obtained using a PtRh (6%)–PtRh (30%) type B thermocouple (diameter 100 μ m). The thermocouple wire was sustained by a fork and crossed the flame horizontally to avoid conduction heat losses. The junction was located at the center of the burner. The thermocouple was coated with an inert layer of BeO – Y_2O_3 to prevent catalytic effects [16]. The ceramic layer was obtained by damping in a hot solution of $Y_2(CO_3)_3$ (93% mass) and BeO (7% mass) followed by drying in a Mecker burner flame. This process was reiterated (about 10 times) until the whole metal was covered. Radiative heat losses were corrected using the electric compensation method [17].

The sampling probe was in silica with a hole of about 50 μ m diameter (d_i). The probe was finished by a small cone with an angle to the vertical of about 20° . For temperature measurements in the flames perturbed by the probe, the distance between the junction of the thermocouple and the end of the probe was taken equal to two times d_i , i.e., about 100 μ m. Gas samples were collected in a Pyrex loop and directly obtained by connecting the quartz probe through a heated line to a volume, which was previously evacuated by a turbomolecular pump down to 10^{-7} kPa and which was then filled up to a pressure of 1.3 kPa (pressures were measured by a MKS 0–100 Torr pressure transducer). The pressure drop between the flame and the inlet of the probe ensure that reactions were frozen. Stable species profiles were determined by gas chromatography. Chromatographs with a Carbosphere packed column and helium or argon as carrier gas were used to analyze O_2 , H_2 , CO , and CO_2 by thermal conductivity detection and CH_4 , C_2H_2 , C_2H_4 , C_2H_6 by flame ionization detection (FID). Heavier hydrocarbons (allene (a- C_3H_4), propyne (p- C_3H_4), cyclopropane (c- C_3H_6), propene (C_3H_6), propane (C_3H_8), vinylacetylene (C_4H_4), butadienes (1,2- C_4H_6 , 1,3- C_4H_6), 1-butyne (1- C_4H_6), 1-butene (1- C_4H_8), isobutene (i- C_4H_8), and benzene (C_6H_6)) were analyzed on a Haysep-packed column by FID and nitrogen as gas carrier gas. The identification of these compounds was performed using GC/MS and by comparison of retention times when injecting the product alone in gas phase. Fig. 1 presents a typical chromatogram of C_3 – C_6 compounds obtained for the flame doped with allene. Contrary to what is observed due to the scale of the figure, the separation between the peaks of isobutene, 1-butene, and 1,3-butadiene and those of vinylacetylene and 1-butyne is acceptable. The peaks

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