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Laminar burning velocities and Markstein lengths of aromatics at elevated temperature and pressure

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Abstract

Laminar burning velocities are presented for benzene, toluene, ethylbenzene, *m*-xylene, and *n*-propylbenzene. The data have been acquired at elevated temperature (450 K) and pressure (304 kPa) over the equivalence ratio range $0.80 \leq \phi \leq 1.4$. High speed schlieren visualization, used to monitor flame growth following ignition, provides a direct determination of the laminar flame speed. The data are corrected for flame stretch to yield the unstretched laminar burning velocities and burned gas Markstein lengths. The data show benzene to be the fastest, followed by ethylbenzene > *n*-propylbenzene > toluene > *m*-xylene. *Iso*-octane results are also presented for comparison to the literature. Flame front perturbations are observed, particularly under fuel-rich conditions, due to hydrodynamic and preferential mass diffusion instabilities. The kinetic factors responsible for the changes in the laminar burning velocities upon alkyl substitution of the aromatic ring are discussed.

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1. Introduction

During the past two decades, the detailed combustion behavior of prototypical aromatics has increasingly been the focus of experimental attention. In contrast to the case of paraffins and oxygenates, for which detailed kinetic mechanisms that reproduce fundamental combustion data have been developed [1,2], an elementary level understanding of aromatic oxidation is only just emerging. The low temperature (~ 600 – 1100 K) reactions of aromatics have been investigated in flow reactor [3–7], rapid compression machine [8–10], and jet-stirred reactor [11–13] studies. Sim-

ilarly, high temperature (~ 1200 – 2600 K) aromatic chemistry has been studied in low pressure laminar flame [14], shock tube [15–20], and diffusion flame studies [21–25]. The studies have helped to shed light on the subtle and complicated chemistry of the aromatic ring destruction as well as substituent effects on reactivity. Nevertheless, fundamental data for important prototypical aromatics remain unavailable.

The laminar burning velocity is an important fundamental property of a fuel that has been the subject of much experimental and modeling interest. On a practical level, it affects the burn rate in internal combustion engines and therefore the engine's efficiency [26,27] and emissions. On a more fundamental level, the burning velocity is an intrinsic property of a combustible mixture, and thus is a useful target for kinetic mechanism validation. Numerous burning velocity measurements

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were carried out for aromatics during the 1950s–1960s using a variety of experimental techniques [28–32], though the results often qualitatively and quantitatively disagreed. It is now recognized that a major source of the discrepancies is the neglect of flame curvature and aerodynamic strain, collectively denoted *stretch* [33–37], which can dramatically alter the burning velocity. Thus, while the earlier data, where consistent, are useful for providing qualitative information on fuel structure effects on burning velocity, they cannot be readily used for validation of detailed kinetic models. While stretch-corrected burning velocity measurements have now been reported for several hydrocarbon fuels, benzene and toluene are the only aromatics for which stretch-corrected data have been reported [24,25]. One of the goals of the present study is to provide stretch-corrected burning velocity measurements for prototypical aromatic fuels to assist validation of elementary chemical kinetic mechanisms. A parallel effort in our laboratories involves the construction of detailed kinetic mechanisms for aromatic fuels [38], and a detailed comparison of these models to the experimental burning velocities measured as part of this work will be presented elsewhere.

Several researchers have shown that an outwardly propagating spherical flame at constant pressure can be used to determine stretch-free burning velocities. The established procedure involves high-speed imaging of the flame front and extrapolation of the measured flame speed to zero-stretch conditions [39,40]. Such an approach is used herein. Recent studies using the constant volume vessel with direct flame visualization [36,39–41] and the counterflow twin opposed flame technique [24,42] show good agreement.

In addition to providing stretch-free laminar burning velocities, direct visualization of spherical flames provides information about flame response to stretch. Stretch effects play an important role in modifying the properties of strongly turbulent flames [43,44] and in controlling flame behavior near extinction [35,45]. For small values of flame curvature and aerodynamic strain, the resulting flame speed is linearly altered from that of the unstretched value according to the relationship

$$s_u = s_u^0 - LK_u, \quad (1)$$

where s_u and s_u^0 are the stretched and unstretched flame speeds, L is the Markstein length, and K_u is the stretch rate. Convenience often dictates normalizing the Markstein length by the characteristic flame thickness δ , usually based on the thickness of the diffusion zone, to yield the Markstein number $Ma = L/\delta$. In the present study, we have not carried out the normalization since consensus has not been reached on accurate procedures for calculating δ .

This analysis procedure is valid for smoothly propagating spherical flames. In the present experimental study, hydrodynamic and preferential diffusion instabilities are identified which alter the flame morphology. The effect of these on the burning velocity and Markstein length determinations are discussed below.

2. Experimental

The experimental apparatus, shown schematically in Fig. 1, has been described previously [26,46] and will only be discussed briefly. The stainless steel vessel has a 16.5 cm diameter spherical cavity (volume = 2.4 L) with four windows for optical access. The two largest are 7.6 cm sapphire windows (6.4 cm clear aperture) arranged on opposite sides of the vessel. These windows allow transmission of a collimated light beam for schlieren diagnostics. Spark electrodes, used as the ignition source for the experiments described herein, are mounted on the top and bottom of the vessel. The vessel is housed in a temperature-controlled oven capable of maintaining a constant temperature to within 2 °C over an extended time period (weeks). The oven has quartz windows to allow transmission of the optical beams.

The gas mixtures are prepared by first charging the hydrocarbon from a vaporizing chamber held at 100 °C to the pressure required for the desired mixture stoichiometry. The stated purity levels of the hydrocarbon fuels are as follows: benzene, toluene, and ethylbenzene—99.8%; *m*- and *p*-xylene—99+%; and *o*-xylene—98%. No further purification was carried out. A high sensitivity MKS Baratron (10,000 Torr range) is used to meter the fuel. The high Baratron sensitivity permits 0.1–0.2 Torr repeatability in the fuel charge,

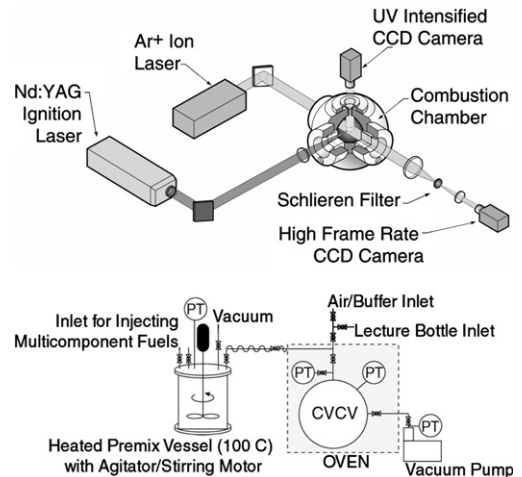


Fig. 1. Schematic of experimental apparatus.

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