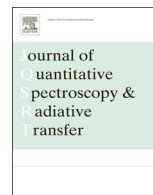




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Laser absorption diagnostic for measuring acetylene concentrations in shock tubes

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ABSTRACT

A fixed-wavelength direct absorption laser diagnostic for high-temperature measurements of acetylene concentration was developed. The diagnostic, based on a tunable continuous wave distributed feedback diode laser, was optimized primarily for studying chemical kinetics behind reflected shock waves. The center wavelength (3335.55 cm^{-1}) of the tunable diagnostic was typically set at the peak of the 3300 cm^{-1} absorption band of acetylene at high temperatures. The absorption spectrum of acetylene diluted in argon was characterized using scanned-wavelength direct absorption measurements from 1070 to 1720 K and 0.8 to 4.0 atm. Line fitting of the measured absorption spectra was not possible due to the large number of transitions overlapped by pressure broadening that contribute to the spectrum. Instead, empirical fits for the peak absorption coefficient and its corresponding wavelength as a function of temperature and pressure were generated. Furthermore, in order to allow for characterization of interference absorption in kinetic studies, empirical fits for the acetylene absorption coefficient in the region around the primary absorption feature were developed. Absorption coefficient measurements of propyne and 1-butyne, which may be the primary interference candidates, reveal that their absorption coefficients are constant in the wavelength range of interest, and are much smaller than those of acetylene. Therefore, the acetylene concentration in the presence of these interfering species can be inferred using two-color techniques. The utility of the acetylene diagnostic was demonstrated by measuring acetylene mole fraction time-histories during the pyrolysis of propene and 1-butene.

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1. Introduction

Acetylene is an important intermediate or product species during the combustion of many hydrocarbon fuels. It is also one of the primary precursors to soot [1]. Improving the experimental tools available for performing kinetic studies of reacting systems involving acetylene is thus of significant interest to the combustion community. Due to their MHz time response and in-situ measurement capabilities,

continuous wave (CW) laser absorption diagnostics have become an invaluable tool for studying chemical kinetics in shock tubes [2–4]. Though measurements of the acetylene mole fraction using chirped- and scanned-wavelength laser absorptions, gas chromatography, and time-of-flight mass spectrometry have already been performed in kinetic studies [5–7], laser absorption diagnostics optimized for high-temperature, high-temporal resolution studies have not yet been demonstrated. In this work, a fixed wavelength direct absorption laser diagnostic for measurements of acetylene concentrations in shock tubes was developed. In addition, the utility of the proposed diagnostic for performing chemical kinetic studies was demonstrated by measuring

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acetylene species time-histories during the pyrolysis of propene and 1-butene.

The IR spectrum of acetylene has been studied in great detail both theoretically and experimentally. Several of these studies have been used to develop the HITRAN 2012 spectroscopic database [8], which contains a comprehensive description of the acetylene spectrum in the 3300 cm^{-1} band that is of primary interest in this work. The acetylene spectrum near this wavelength is primarily composed of two cold bands, the ν_3 band and the $\nu_2+(\nu_4+\nu_5)^0$ combination band, as well as at least 18 hot bands [9]. Line positions and intensities for the two cold bands in the HITRAN 2012 database [8] were taken from work by Auwera et al. [10], and air- and self-broadening coefficients were taken from work by Devi et al. [11] and Varanasi et al. [12]. Details on the spectral parameters for the hot bands are described by Jacquemart et al. [9]. Though significant effort has been made to include hot transitions in the HITRAN 2012 database [8], recently measured emission spectra by Moudens et al. [13] demonstrate that the database does not account for several hot-band transitions involving highly excited vibrational levels. Indeed, this is confirmed experimentally in this work and is discussed in Section 4.

2. Experimental methods

The spectral location of the proposed acetylene diagnostic lies at the peak of the 3300 cm^{-1} absorption band, as shown in Fig. 1. Though acetylene exhibits stronger absorption near 700 cm^{-1} , this wavelength is not easily accessible using current lasers. Furthermore, the 3300 cm^{-1} band offers stronger absorption compared to the 1300 cm^{-1} and 6500 cm^{-1} bands which were used to perform laser absorption measurements of acetylene in previous studies [6,7]. Although simulations using the HITRAN 2012 database [8] do not fully agree with experimental measurements (see Section 4), the authors believe that the database is sufficiently accurate for selecting the optimal wavelength for the proposed diagnostic.

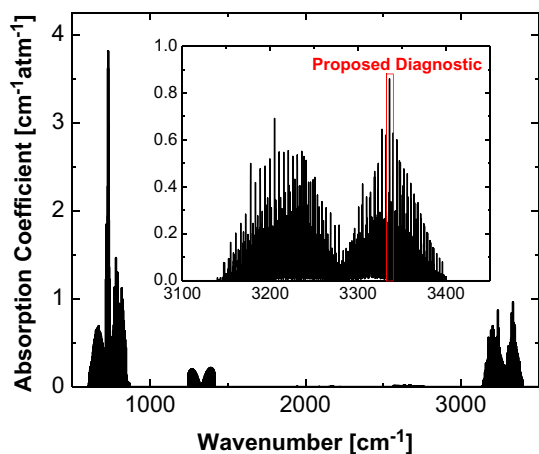


Fig. 1. Absorption spectrum of acetylene at 1400 K, 1 atm calculated using HITRAN 2012. Primary plot shows the entire spectrum from 500 to 3500 cm^{-1} , subplot shows spectrum in the 3300 cm^{-1} band.

The absorption spectrum of acetylene was measured using scanned-wavelength direct absorption (DA) [14] at room temperature in a 79.9 cm pathlength cell, and at high temperatures behind reflected shock waves in a shock tube. The 14.13 cm diameter shock tube used in this study is described in previous work [15–17]. Measurements at room temperature and pressure were performed in order to characterize the performance of the laser (Nanoplus DFB laser, $\lambda=2998\text{ nm}$ @ $30\text{ }^\circ\text{C}$) and detector systems (Vigo Systems PVI-3TE-4), and to validate simulations using the HITRAN 2012 database [8] at these conditions. Scanned-wavelength DA measurements were performed from 3335.1 cm^{-1} to 3335.9 cm^{-1} using a sawtooth signal scanned at 1 and 2.5 kHz in the static cell and shock tube, respectively, with a peak-to-peak modulation current ranging from 200 to 350 mA ($dl/dv=-38\text{ mA/cm}^{-1}$). The ideal constant-volume (CV) test time in the shock tube was 1 ms, which allowed for at least 2 full scans of the acetylene spectrum per experiment. All shock tube measurements were performed using 2% acetylene/argon mixtures purchased from Praxair (acetone free).

The absolute wavelength of the peak of the acetylene absorption feature at room temperature and pressure was measured using a Bristol 721 wavelength meter ($\nu_{\text{uncertainty}}=\pm 0.0035\text{ cm}^{-1}$). The relative wavelength in scanned-wavelength experiments was determined by measuring the transmission peak spacing of a solid germanium Fabry–Perot etalon ($\text{FSR}=0.0162\text{ cm}^{-1}$). The wavelength shift of the peak of the acetylene absorption feature relative to that at room temperature and pressure was determined by simultaneously measuring the laser intensity of a secondary beam that was pitched through a 2.5 cm reference cell filled with a 0.2% acetylene/argon mixture. Since knowledge of the absolute wavelength is not critical during the implementation of the diagnostic in kinetic studies, the authors recommend centering the laser at the peak of the acetylene absorption feature at the experimental conditions by adjusting the laser injection current/wavelength relative to the absorption peak of acetylene in the reference cell. This laser centering method requires accurate knowledge of the wavelength shift of the peak absorption coefficient at the experimental conditions relative to that at room temperature and pressure, as well as a precise determination of the injection current-to-wavelength relationship for the laser device, both of which were measured accurately in this work. The uncertainty in the relative wavelength using this method is no greater than $\pm 0.002\text{ cm}^{-1}$, which is significantly more accurate than absolute wavelength measurements using most commercial wavelength meters.

The schematic for the proposed acetylene diagnostic for use in kinetic studies is shown in Fig. 2. Due to the sub-0.15% low- and high-frequency noises of the laser/detector system, normalizing the measured transmitted laser intensity through the shock tube by the measured laser intensity at a reference detector upstream of the shock tube (common-mode-rejection) was not necessary. However, due to the relatively low power ($\sim 3\text{ mW}$) of the laser used in this work, measurements indicated that light emission due to acetylene or other hydrocarbons might affect laser absorption measurements. Therefore, as shown in Fig. 2, an aperture and a narrow bandpass filter ($\text{CWL}=3332\text{ cm}^{-1}$, $\text{FWHM}=35\text{ cm}^{-1}$) were

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