Combustion and Flame 163 (2016) 90–99

Contents lists available at ScienceDirect

Combustion and Flame

journal homepage: www.elsevier.com/locate/combustflame

Combustion kinetics of laser irradiated porous graphite from imaging Fourier transform spectroscopy

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ARTICLE INFO

Article history:
Received 6 April 2015
Revised 3 September 2015
Accepted 6 September 2015
Available online 14 November 2015

Keywords:
Graphite
Laser heating
Combustion plume kinetics
Hyperspectral imaging
Laser lethality

ABSTRACT

The combustion plumes arising from laser-irradiated graphite targets were investigated experimentally using hyperspectral, imaging Fourier transform spectroscopy (IFTS). Porous graphite targets were irradiated with a 1.07 μm, 20-kW ytterbium fiber laser at irradiances of 0.25–4 kW/cm^2. Emissive plumes from the oxidation of graphite in air were monitored using a mid-wave infrared imaging Fourier-transform spectrometer with spatial resolution of 0.52 mm per pixel. Strong spectral emission of CO and CO_2 were observed in the infrared between 1900 and 2400 cm^{-1} with an instrument spectral resolution of 2 cm^{-1}. A homogeneous single-layer plume, line-by-line radiative transfer model (LBLRTM) and two band models (EM2C and RADCAL) were applied to estimate spatial maps of temperature and column densities of CO and CO_2 with a temporal resolution of 0.47 s per hyperspectral image. Steady surface temperatures of 1800–2900 K are achieved after ~1 min for irradiances of 0.25–1.0 kW/cm^2. A stable, gas phase combustion layer extends from 4 to 12 mm from the surface, with buoyancy driving a gas flow of ~8 m/s. Plume extent and intensity is greater for the larger porosity (6 mm particle size) samples. Steady-state gas temperatures exceed surface temperature by up to 400 K. Column densities for CO and CO_2 of up to 10^{18} molec/cm^2 were observed. The CO/CO_2 concentration ratio peaks at 2500 K. The initial rise with temperature is consistent with effective activation energies of 149–111 kJ/mol at distances between 0.72 mm and 3 mm, respectively.

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1. Introduction

The development of high energy lasers for weapons applications has been pursued for more than 50 years, including the Airborne Laser for missile defense and the Tactical High Energy Laser for counter rocket, artillery and mortar applications [1]. Weapon effectiveness depends on many parameters including system performance (laser power, wavelength, spot size), engagement conditions (range, dwell time, atmospheric conditions), and target vulnerability (aim point, lethal fluence, structural loading). With the new focus on tactical missions, the advent of high power fiber lasers at shorter wavelengths, and targets with new composite materials, the need for understanding laser-material interactions is growing dramatically. The pyrolysis and subsequent gas phase combustion of carbon rich composite materials is of particular interest [2–5]. Combusting gas and particulate plumes above the surface can significantly alter the laser energy coupled to the target.

Imaging Fourier transform spectroscopy (IFTS) with high framing rates is emerging as an important tool for monitoring combustion processes [3,6–8]. We are developing a mid-infrared IFTS system with spatial resolution of up to 0.5 mm over a 256 × 320 array, spectral resolution of up to 0.25 cm^{-1} and hyperspectral data cube framing rates of 0.1–10 Hz for combustion event monitoring [3,6–8]. Two-dimensional imaging of combustion plumes above laser-irradiated samples would be particularly useful in developing profiles for molecular species concentrations and gas temperatures in the boundary layer, benchmarking reactive fluid dynamic calculations. In the present work, we develop spatial maps of combustion products above laser irradiated graphite targets. High heating rates yield surface temperatures of up to 4000 K and buoyant boundary layers with thickness of > 1 cm.

The global heterogeneous production of CO and CO_2 from graphite surfaces is often characterized by the partitioning [9–12]:

$$\frac{[CO]}{[CO_2]} = Ae^{-B/T} \tag{1}$$

where the pre-exponential and activation temperature parameters, A and B, depend significantly on the carbon source and reactor design, and temperatures are typically limited to below 1800 K [11]. There is some controversy regarding the direct production of CO_2 from the oxidation of a graphite surface, due to the subsequent homogeneous conversion of CO to CO_2, the surface reaction of CO_2 to regenerate the CO, and the role of diffusion [9–18]. The general consensus is that the

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http://dx.doi.org/10.1016/j.combustflame.2015.09.004
0010-2180/Published by Elsevier Inc. on behalf of The Combustion Institute.

direct CO_2 production is small for elevated temperatures [12,19–21]. However, the surface oxidation mechanism does include elementary steps for the production of both CO and CO_2 [22]. Semi-global rate packages for the heterogeneous reactions for both porous and non-porous graphite have been developed [19–21]. The current work evaluates the relationship in Eq. (1) at temperatures up to 3000 K.

There is a strong coupling between the homogeneous and heterogeneous kinetics that depends on temperature, flow rate, oxidizer, and pressure [20,21]. The effect of particle size on diffusion rates has also been studied in some detail [20–23]. To simplify the diffusion and fluid dynamics in fluidized bed reactors, analytic expressions for the $[CO/[CO_2]$ ratio have been developed using single and double layer films [10,24,25]. The concentration ratio initially increases with temperature, but can exhibit a peak near 1100 K for combustion in moist air, depending on particle size. We seek to evaluate these predictions for the elevated temperatures and different diffusion conditions encountered in these high power laser based experiments.

2. Experimental

Porous graphite targets from Graphtek of 7.62 × 7.62 cm^2 and 1.27 or 0.64 cm thickness, were irradiated by a 20 kW IPG Photonics model YLR-2000 ytterbium fiber laser at 1.07 μm, as shown in Fig. 1. Pyrolytic graphite with density of 2.20 g/cm^3 and two extruded graphite samples with density of 1.55 g/cm^3 (course porosity with typical particle diameter of 0.6 cm) and 1.72 g/cm^3 (medium porosity with particle diameter of 0.15 cm) were irradiated at normal incidence in air with buoyancy and a distant fume hood driving a slow vertical gas velocity of ~8 m/s. The sample holder was a vise clamp insulated with ceramic between the sample and the steel serrated jaws, holding the sample at the bottom edge. A near Gaussian beam with a one-sigma radius of 2.23 cm and irradiances of 0.25–4 kW/cm^2 were incident on the graphite samples for 120 s.

The primary optical diagnostic is a Telops MWIR (1800–6667 cm^{-1}) imaging Fourier transform spectrometer (IFTS) viewing the gas plume evolving from the surface. The IFTS was placed at a focal distance of 47 cm from the center of the plume and observes hot gas emission integrated along the line of sight (*z*-axis) with two-dimensional image away from surface horizontally (*x*-axis) and along the surface vertically (*y*-axis). A large cold blackbody is placed on the far side of the plume to provide a uniform background. The InSb focal plane array (FPA) was narrowed to 200 × 64 pixels with a spatial resolution of 0.52 mm/pixel. Interferograms were recorded on each pixel with a spectral resolution of 2 cm^{-1}. The Fourier-transform of the interferograms produces full hyperspectral data cubes with a temporal resolution of 0.47 s.

The IFTS is calibrated using two wide area blackbodies at 673 and 873 K to determine the gain and offset necessary to convert the raw spectra to spectral radiance (W/cm^2 sr cm^{-1}) [26]. The 16-bit FPA using a 0.3 optical density neutral density (ND) filter saturates with 65,000 counts at 300 $\mu W/(cm^2$ sr $cm^{-1})$, with a background radiance of 6 $\mu W/(cm^2$ sr $cm^{-1})$. For gas plume temperatures near 1000 K, the DC component of the interferogram represents 28 % of the dynamic range, with the interferometer producing an 8% modulation at zero optical path difference (ZPD) [3]. For the hotter plumes encountered in this work, a 2.0 ND filter is used. The plume radiance at 2900 K yields a filtered irradiance of 1000 $\mu W/(cm^2$ sr $cm^{-1})$. The ratio of the DC component to center burst for these conditions is 72%.

A Phantom v7 three-color, CMOS visible camera with 1024 × 1024 pixels provides full frame imagery at 350 Hz. At a distance of 72 cm, the individual pixel field of view is 0.14 mm/pixel. The visible imagery provides information on flow field, including ejected particle sizes and velocities. Surface temperature was monitored with a FAR Associates Spectro-Pyrometer with spatial averaging over the central 0.75 cm radius of the irradiated spot, or the central 33% of the Gaussian beam diameter. Further details regarding the experimental apparatus and procedures have previously been documented [8].

3. Results

3.1. Plume dynamics

Images of the infrared emissive plumes at several irradiances levels for the coarser porosity samples are shown in Fig. 2. A temporal average of 1.2 × 10^6 interference images over the full 120 s irradiation produce broadband images that define the spatial extent and structure of the combustion products. By temporally averaging the inteferograms, the instrument modulation and scene change artifacts are largely eliminated and only the DC component associated with the broadband image is retained [27,28]. Blackbody emission from the hot 1.27 cm graphite sample located in the bottom left corner of each image was masked to eliminate saturated pixels. The laser beam is centered on the sample at $y = 33.12$ mm and the Gaussian width extends from $y = 10.08$ to 54.72 mm. Combustion is weak for the laser irradiance of $I = 251$ W/cm^2, where the surface temperature is $T_s = 1800$ K and the heterogeneous oxidation is relatively slow. As the laser irradiance is increased from 485 W/cm^2 to 1 kW/cm^2 the surface temperature increases from 2200 K to 2900 K. Under these

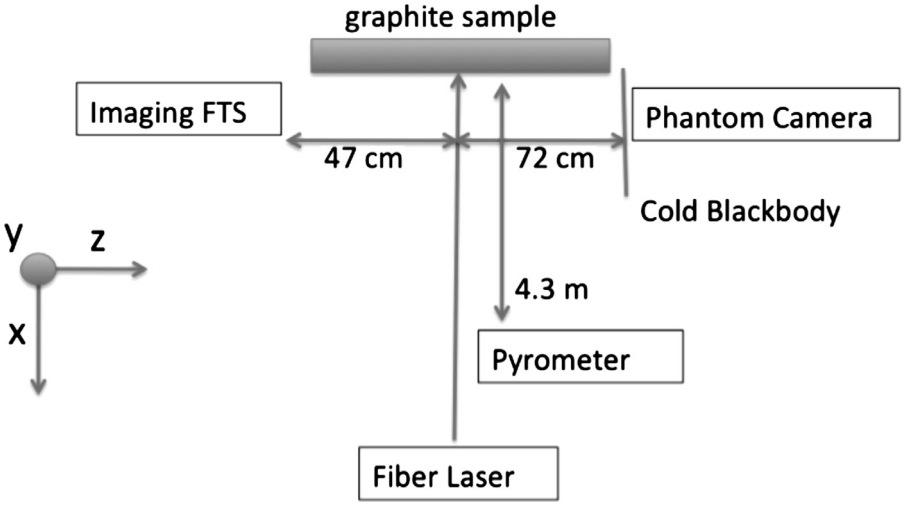

Fig. 1. Experimental apparatus.