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journal homepage: www.elsevier.com/locate/hmtImpacts of N₂ and CO₂ diluent gas composition on flame emission spectroscopy for fuel concentration measurements in flamesSungkyun Oh^{a,b}, Youchan Park^{a,b}, Guwon Seon^a, Wontae Hwang^{a,b}, Hyungrok Do^{a,b,*}^a Department of Mechanical and Aerospace Engineering, Seoul National University, Seoul 08826, South Korea^b Institute of Advanced Machines and Design (IAMD), Seoul National University, Seoul 08826, South Korea

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ABSTRACT

Flame emission spectroscopy (FES) is used to determine methane fuel concentration in a premixed flat-flame burner with varying diluent composition. Spectral profiles of the molecular emission bands of OH* (306.4 nm), CH* (431.5 nm), and C₂* (520 nm) in the UV-to-visible range are recorded and analyzed for various equivalence ratios ($0.72 < \phi < 1.25$), oxygen concentration ($0.19 < X_{O_2} < 0.35$), and diluent gas species (N₂, CO₂). The importance of flame radiation, which is strong and reliable in the UV spectrum, is confirmed particularly for quantitative measurement purposes. In air-methane flames, the band intensity ratios of OH*/CH* and C₂*/CH* are shown to be highly sensitive to fuel stoichiometry, and can thus serve as accurate indicators of ϕ . In addition, the impact of diluent gas composition on flame radiation characteristics and FES utilizing band intensity ratios is investigated for potential applications such as clean combustion utilizing exhaust gas recirculation (EGR), and oxy-combustion in supercritical CO₂ power plants. It is shown that the strength of the underlying broadband emission is strongly dependent on the diluent gas composition, and influences the sensitivity and accuracy of the FES-based ϕ measurement.

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1. Introduction

Measuring flow properties in combustion environments is essential for controlling combustion heat release and efficiently operating propulsion and power generation systems. In particular, fuel/air (FA) ratio is one of the most important parameters that directly impact flame temperature, combustion dynamics, and emission characteristics. For measuring FA ratio, devices such as flame ionization detectors (FID), potentiometric FA sensors, and gas chromatograph (GC) have been introduced in hydrocarbon-fueled combustion systems. Alternatively, non-intrusive in-situ optical measurement methods have been proposed and developed that do not perturb and interact with the reacting flow. In order to induce photon-matter interactions in the target flows, the optical techniques typically require photon sources that have different power levels: low power for absorption spectroscopy and high power for laser-induced fluorescence (LIF), Raman scattering, and laser-induced breakdown spectroscopy (LIBS). On the other hand, no external photon source is required for measurements utilizing gas-phase flame radiation, such as flame emission spectroscopy (FES),

which is a passive but reliable way of measuring local gas properties during combustion.

When sufficient heat energy is released from the reaction zone, the flame radiation (chemiluminescence) is due to the spontaneous emission of excited intermediate and product species. For instance, electronically excited CH* emits photon energy at 387.2 and 431.5 nm, after collisional excitation and subsequent chemical reactions of $CH_2 + O \rightarrow CO + CH^*$ and $C_2H + O_2 \rightarrow CO_2 + CH^*$ [1]. In addition, quasi-continuum CO₂* emission appears in a broad spectral range from ultra-violet (UV) to visible wavelengths [2,3]. Therefore, the overall flame radiation is composed of both broadband CO₂* emission and discretized molecular band spectra, where the position and strength of each line, band, and system are determined by the chemical kinetics associated with the hydrocarbon combustion reactions and interacting flow properties. This implies that the chemiluminescence contains useful information regarding the gas composition and flow properties such as density and temperature.

The emission spectra of typical combustion intermediate and product species in the UV to visible range, such as CH* (431.5 nm), OH* (306.4 nm), C₂* (517 nm), CN* (350 nm), and CO₂* (250–800 nm) bands, have been extensively investigated and well-tabulated. These bands strongly depend on temperature and density conditions [3–6]. It has been demonstrated that the flame tem-

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