



# The effect of oxygen enrichment on soot formation and thermal radiation in turbulent, non-premixed methane flames

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## Abstract

Non-premixed oxy-fuel combustion of natural gas is used in industrial applications where high-intensity heat is required, such as glass manufacturing and metal forging and shaping. In these applications, the high flame temperatures achieved by oxy-fuel combustion increase radiative heat transfer to the surfaces of interest and soot formation within the flame is desired for further augmentation of radiation. However, the high cost of cryogenic air separation has limited the penetration of oxy-fuel combustion technologies. New approaches to air separation are being developed that may reduce oxygen production costs, but only for intermediate levels of oxygen enrichment of air. To determine the influence of oxygen enrichment on soot formation and radiation, we developed a non-premixed coannular burner in which oxygen concentrations and oxidizer flow rates can be independently varied, to distinguish the effects of turbulent mixing intensity from oxygen enrichment on soot formation and flame radiation. Local radiation intensities, soot concentrations, and soot temperatures have been measured using a thin-film thermopile, planar laser-induced incandescence (LII), and two-color imaging pyrometry, respectively. The measurements show that soot formation increases as the oxygen concentration decreases from 100% to 50%, helping to moderate a decrease in overall flame radiation. An increase in turbulence intensity has a marked effect on flame height, soot formation and thermal radiation, leading to decreases in all of these. The soot temperature decreases with a decrease in the oxygen concentration and increases with an increase in turbulent mixing intensity. Overall, the results suggest that properly designed oxygen-enriched burners that enhance soot formation for intermediate levels of oxygen purity may be able to achieve thermal radiation intensities as high as 85% of traditional oxy-fuel burners utilizing high-purity oxygen.

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**Keywords:** Oxy-fuel; Flame; Methane; Soot; Radiation

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## 1. Introduction

Oxy-fuel combustion is a well-known approach for improving the thermal efficiency of high-temperature industrial furnaces [1–3]. The efficiency improvement results from the greater radiant emission from oxy-fuel flames, a consequence

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of the higher flame temperatures and greater emissivity of the product gases. The peak flame temperature during pure oxy-fuel combustion of natural gas is nearly 3000 K, compared to a peak temperature during air-firing of about 2200 K. Absent the dilution from  $N_2$  associated with air-based combustion, the concentrations of  $CO_2$  and steam (both of which are radiantly active) in the products of oxy-fuel flames are 3.5 times greater than in air-fuel flames. As a result, up to 98% of the heat transfer from oxy-fuel flames to the ‘batch’ (the solid particle components that will make up the glass) in glass-melting furnaces has been shown to be through radiation [1]. Overall, glass melting furnaces and steel heating furnaces have shown 20–60% improvement in furnace efficiency when retrofitting from air-fired to oxy-fired operation [3–5]. However, producing oxygen through air separation consumes a large amount of energy, such that the overall system efficiency when switching to oxy-fuel firing may or may not improve, depending on the original furnace efficiency and the energy efficiency of oxygen production.

The penetration of oxy-fuel technology into industrial markets is currently constrained by the high cost of commercial air separation technologies. Cryogenic air separation is the most widely used technology for generating oxygen at a large scale [6,7]. This approach generates oxygen with a purity of 90–99.5% and can scale to large single-unit production sizes. Unfortunately, it is a complex and expensive technology. Pressure swing adsorption (PSA) is a competing technology for relatively small-scale applications that do not require oxygen purity greater than 94%. Current PSA systems rely on preferential adsorption of  $N_2$  onto zeolites and consist of a multi-step batch production process [6–8].

New advances in materials science offer the prospect of lower-cost production of oxygen-enriched air via membranes (e.g. ion-transport membranes [9,10]) or through adsorption approaches with novel materials. Recently, a new class of materials known as metal–organic frameworks (MOFs) has been developed, featuring large surface areas and open metal coordination sites for gas adsorption [11]. Some preliminary studies of MOFs for air separation have been conducted, and recent work at Sandia National Laboratories has identified promising MOFs structures and compositions for improved air separation [12,13]. In the future, the use of MOFs or other advanced materials may allow cost-effective oxygen production at near-ambient temperatures, but with an imperfect purity. If carbon capture from the exhaust is desired, having a high-purity oxygen stream is of paramount importance.

Within this context, the question arises as to the level of oxygen enrichment necessary to achieve the desired heat transfer in industrial oxy-fuel applications. Specifically, one would like to know how

much enhancement in radiative heating is possible with different levels of oxygen enrichment, and how oxygen-enriched burners can be designed to maximize radiant emission. The presence of soot within a flame is known to enhance radiant heat transfer, potentially to a significant extent [14–16]. The relationship between turbulent mixing, soot formation, and flame radiation in oxygen-enriched methane flames has not been previously explored in depth. Baukal and Gebhart [17] varied  $O_2$  levels from 28% to 100% in a rapid mixing burner firing natural gas from laminar to turbulent flow and across a range of overall stoichiometries. They measured “non-luminous” thermal radiation (i.e. without soot contributions) and found it to increase rapidly with oxygen purity, with only a small tapering of the rate of increase as the oxidizer composition approached the pure oxygen limit. Wang et al. [18] studied mean soot formation and thermal radiation from a series of natural gas and propane flames formed by injecting turbulent fuel jets into low-velocity laminar oxidizer streams consisting of 21–100%  $O_2$ . They found a peak soot volume fraction for 70%  $O_2$  when using a modestly turbulent fuel jet ( $Re = 5000$ ) and a peak soot volume fraction for 35%  $O_2$  when using a more strongly turbulent fuel jet ( $Re = 15,000$ ).

Here, we expand upon these previous studies by utilizing more realistic turbulent flows of both methane and oxidizer and by keeping the stoichiometry of supplied burner gases near one, as is practically relevant. We report detailed, spatially resolved measurements of both radiation and soot concentrations, as well as mean soot temperature in these flames.

## 2. Experimental description

### 2.1. Turbulent co-annular burner

For this investigation a co-annular burner geometry was selected in which the fuel is emitted from a 4.76 mm outer diameter (OD) smooth, seamless stainless steel tube that is surrounded by a 12.70 mm OD smooth, seamless stainless steel tube in which the oxidizer flows (see Fig. 1). The internal diameter (ID) of the fuel tube is 3.34 mm and that of the oxidizer tube is 10.21 mm. Both tubes have straight-cut ends to facilitate flame holding, with the fuel tube penetrating 1 mm beyond the oxidizer tube. The fuel and oxidizer tubes have lengths of 80 cm and 55 cm, respectively. Wire spacers recessed 140 mm from the burner tip are used to centralize the fuel tube. The theoretical length needed for fully developed turbulent pipe flow under our operating conditions ranges from 94 to 106 mm. Similarly, the fuel tube is four times longer than required to form fully developed pipe flow. Outside of the oxidizer tube, a well-conditioned 800 slpm coflow of air or  $N_2$  was supplied in an 18.7 cm circular duct, giving an exit velocity of 0.49 m/s, to shield the flame from

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