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Characteristics of non-premixed oxygen-enhanced combustion: I. The presence of appreciable oxygen at the location of maximum temperature

S.A. Skeen [a], G. Yablonsky [a,b], R.L. Axelbaum [a,*]

[a] *Department of Energy, Environmental, and Chemical Engineering, Washington University in St. Louis, One Brookings Drive, Box 1180, St. Louis, MO 63130, United States*
[b] *Parks College, Saint Louis University, St. Louis, MO 63103, United States*

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ABSTRACT

The presence of appreciable molecular oxygen at the location of maximum temperature has been observed in non-premixed oxygen-enhanced combustion (OEC) processes, specifically in flames having a high stoichiometric mixture fraction (Z_{st}) produced with diluted fuel and oxygen-enrichment. For conventional fuel–air flames, key features of the flame are consistent with the flame sheet approximation (FSA). In particular, the depletion of O_2 at the location of maximum temperature predicted by the FSA correlates well with the near-zero O_2 concentration measured at this location for conventional fuel–air flames. In contradistinction, computational analysis with detailed kinetics demonstrates that for OEC flames at high Z_{st}: (1) there is an appreciable concentration of O_2 at the location of maximum temperature and (2) the maximum temperature is not coincident with the location of global stoichiometry, O_2 depletion, or maximum heat release. We investigate these phenomena computationally in three non-premixed ethylene flames at low, moderate, and high Z_{st}, but with equivalent adiabatic flame temperatures. Results demonstrate that the location of O_2 depletion occurs in the vicinity of global stoichiometry for flames of any Z_{st} and that the presence of appreciable O_2 at the location of maximum temperature for high Z_{st} flames is caused by a shift in the location of maximum temperature relative to the location of O_2 depletion. This shifting is attributed to: (1) finite-rate multi-step chemistry resulting in exothermic heat release that is displaced from the location of O_2 depletion and (2) the relative location of the heat release region with respect to the fuel and oxidizer boundaries in mixture fraction space. A method of superposition involving a variation of the flame sheet approximation with two heat sources is shown to be sufficient in explaining this phenomenon.

1. Introduction

In general, oxygen-enhanced combustion (OEC) refers to the use of elevated oxygen concentrations (>21% by volume) in the oxidizer stream. The benefits of OEC over conventional air-fired combustion include increased temperature and thermal efficiency for industrial heating and melting processes, reduced pollutant emissions and fuel consumption, and improved flame stability [1]. Costs associated with air-separation have limited the number of industrial scale OEC systems; however, recent advances in air-separation technology, rising fuel prices, stricter government emissions standards, and the need to reduce atmospheric carbon dioxide (CO_2) emissions have created an economic environment where the costs of OEC have diminished substantially relative to the anticipated benefits.

For example, Chatel-Pelage et al. [2] reported that current NOx reducing combustion modifications such as the use of overfire air and low-NOx burners will not be able to meet future NOx emissions standards without the addition of selective catalytic reduction technology. Depending on the size and age of the power plant, retrofitting a system to include selective catalytic reduction may be both expensive and difficult. Thus, novel low-NOx combustion modification technologies that would enable existing power plants to meet future government regulations are highly desirable. Several studies have demonstrated that OEC alone, and coupled with unique burner design and oxygen placement, can reduce NOx emissions beyond levels achieved by using overfire air and low-NOx burners [3–11].

Furthermore, rising atmospheric CO_2 concentrations and their potential link to global climate change have generated enormous interest in the development of technologies to reduce CO_2 emissions from combustion processes. Oxy-fuel combustion, which involves replacing the oxidizer air with a mixture of oxygen and externally recycled flue gases, has been identified as an enabling technology for CO_2 capture from coal-fired power plants. Under conventional air-fired conditions the exhaust from a coal power plant contains 10–20 vol.% CO_2, requiring expensive amine absorp-

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[*] Corresponding author. Fax: +1 314 935 7211.
E-mail address: axelbaum@wustl.edu (R.L. Axelbaum).

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tion techniques to capture CO_2; however, studies have demonstrated that oxy-coal power plants can achieve up to 95 vol.% CO_2 in the exhaust enabling CO_2 capture by more direct methods while reducing the costs associated with the cleanup of other pollutant emissions [12].

When utilizing OEC in non-premixed systems the concentrations of the inlet fuel and oxidizer can have a dramatic impact on the flame structure, i.e. the relationship between local temperature and local species concentrations [13–27]. When OEC is combined with fuel-dilution such that a constant amount of inert is present at the flame front, the adiabatic flame temperature is unchanged. Nonetheless, the flame structure is changed dramatically [16]. The extent of oxygen-enrichment and fuel-dilution can be quantified by the stoichiometric mixture fraction, Z_{st}, given by

$$Z_{st} = (1 + \sigma Y_{F,fu}/Y_{O_2,ox})^{-1} \tag{1}$$

where $\sigma = \upsilon_{O2}W_{O2}/\upsilon_F W_F$ and Y_i, υ_i, W_i represent the mass fraction, stoichiometric coefficient, and molecular weight of species *i*, respectively. The fuel stream boundary is indicated by the subscript *fu* and the oxidizer stream boundary is indicated by the subscript *ox*. Thus, pure fuel burning in air represents a low Z_{st} flame while heavily diluted fuel burning in pure oxygen represents a high Z_{st} flame.

Several studies have examined the effects of varying Z_{st} on the flame structure and combustion characteristics for gaseous fuels [13–27]. Many of these studies [14–22,24,26] have demonstrated that increasing Z_{st} has an inhibitory effect on soot formation, while others have demonstrated improved flame strength [23,27]. Sugiyama [14] and Lin and Faeth [15] concluded that the soot inhibiting effect of oxygen-enrichment and fuel-dilution was due to hydrodynamic effects while Du and Axelbaum [16] proposed that soot formation was reduced at high Z_{st} due to changes in flame structure resulting in less fuel and more oxygen in the region of high temperature. Furthermore, using a flame code with detailed chemistry Du and Axelbaum showed that increasing Z_{st} in ethylene flames can result in a nearly two orders of magnitude increase in the concentration of molecular oxygen at the location of maximum temperature ($x_{T\max}$), with molar concentrations of O_2 reaching 8 vol.%. This phenomenon is referred to here as *appreciable molecular oxygen at the location of maximum temperature* and has been subsequently observed both numerically and experimentally by Sun et al. [19] in acetylene flames and Cheng et al. [25] in methane flames. Experimental results suggest that characteristics of the OEC-process are quite sensitive to changes in the concentration of molecular oxygen at the location of maximum temperature.

With regards to flame strength, Du and Axelbaum [13], Chen and Axelbaum [23], and Kitajima et al. [27] showed experimentally that increasing Z_{st} results in a higher strain rate and scalar dissipation rate at extinction for non-premixed counterflow flames. At high Z_{st} extinction scalar dissipation rates 20–40 times greater than for fuel–air flames can be obtained even when the equilibrium flame temperatures are the same. To understand their results Du and Axelbaum and Chen and Axelbaum simulated the flames using a counterflow flame code and proposed that the availability of oxygen in the high temperature region due to increased Z_{st} accelerates the chain branching reaction $H + O_2 = OH + O$ resulting in improved flame strength. Furthermore, Chen and Axelbaum [23] found that at high Z_{st} the flames are able to resist extinction at lower temperatures because at high Z_{st} the location of radical production and $x_{T\max}$ are coincident, which allows the branching reaction to “make most efficient use” of the high temperature zone. On the other hand, in fuel–air flames the radical production zone is found at a lower temperature on the oxidizer side of $x_{T\max}$.

The increased presence of oxygen and oxidizing species in the region of high temperature for high Z_{st} flames can also influence soot inception by affecting both the rate of soot precursor oxidation and the relative location of precursor oxidation to precursor formation. Sunderland et al. [22] proposed a theory suggesting that in order for soot to form in a non-premixed flame the local temperature and local carbon-to-oxygen ratio (C/O) must be above threshold values. Kumfer et al. [26] expanded this theory to include the effects of finite-rate chemistry and residence time and developed a simple model that describes the region conducive to soot inception as being confined between two boundaries: a low temperature boundary that is governed by finite-rate chemistry and can be manipulated by changing the fuel concentration, flame temperature, or characteristic residence time, and a high temperature boundary that is related to the presence of oxygen and oxidizing species and is characterized by the local C/O ratio as described in Sunderland et al. [22].

The above findings indicate that high Z_{st} dramatically affects flame properties, and that one of the primary reasons for this is that high Z_{st} flames have appreciable molecular oxygen at the location of maximum temperature. While the existence of appreciable molecular oxygen has been observed both numerically and experimentally [16,19,25], no explanation has been given in the literature for this phenomenon. The purpose of this work is to develop such an explanation by using a counterflow flame code with detailed chemistry in order to distinguish the key sub-mechanisms responsible for the existence of appreciable O_2 at $x_{T\max}$. Results obtained in this study will further our fundamental understanding of the effect of oxygen-enhancement and fuel-dilution (i.e. increasing Z_{st}) on flame structure.

2. Numerical

The counterflow flame code employed here was originally developed by Kee et al. [28] for premixed counter flow flames and was later modified for adiabatic non-premixed combustion by Lutz et al. [29]. Chemical reaction rates, transport properties, and thermodynamic properties are evaluated by the Chemkin software package. The detailed kinetic mechanism used here consists of 101 species and 544 reactions and was proposed by Wang and Frenklach [30] and modified by Appel et al. [31].

The accuracy of a mechanism similar to that used here was demonstrated by Sun et al. [19] at low and high Z_{st} by comparing numerical results with temperature and species profiles obtained via spontaneous Raman spectroscopy. The slightly larger mechanism used here was validated by modeling the flame conditions of Sun et al. and verifying that temperature and species profiles were replicated.

The computational grid was initialized with 21 grid points and the fuel and oxidizer stream inlet boundaries were spaced 2 cm apart. Adaptive grid refinement was utilized resulting in approx. one-hundred and five grid points on average at convergence. The fuel and oxidizer stream exit velocities were held constant at 80 cm/s for all flames resulting in a strain-rate, *a*, of 62 s^{-1} for the fuel–air case (Flame A), 61 s^{-1} for the $Z_{st} = 0.4$ case (Flame B), and 68 s^{-1} for the $Z_{st} = 0.78$ case (Flame C). The strain rates were determined by fitting a straight line to the computed velocity profile upstream of the thermal mixing layer on the oxidizer side for Flames A and B, and on the fuel side for Flame C. Chen and Axelbaum [23] reported numerically determined extinction strain rates greater than 2400 s^{-1} for ethylene counterflow flames at $T_{ad} = 2370$ K, thus the flames considered in this study are far from extinction. The square of the gradient in mixture fraction $(dZ/dx)^2$, which is proportional to the scalar dissipation rate, was also calculated at the location of stoichiometry for all flames. The values were found to be 0.45 cm^{-2}, 5.76 cm^{-2}, and 4.12 cm^{-2} for Flames A, B, and C, respectively. To ensure that the higher scalar dissipation rates of Flames B and C were not responsible for the increasing