COMBUSTION RESEARCH FACILITY

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The Effect of Oxygen Enrichment on Soot Formation and Thermal Radiation in Turbulent, Non-Premixed Methane Flames

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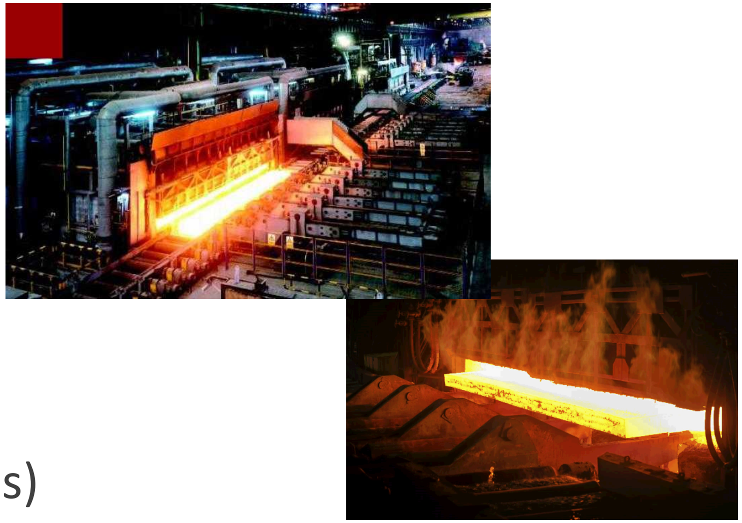
Background

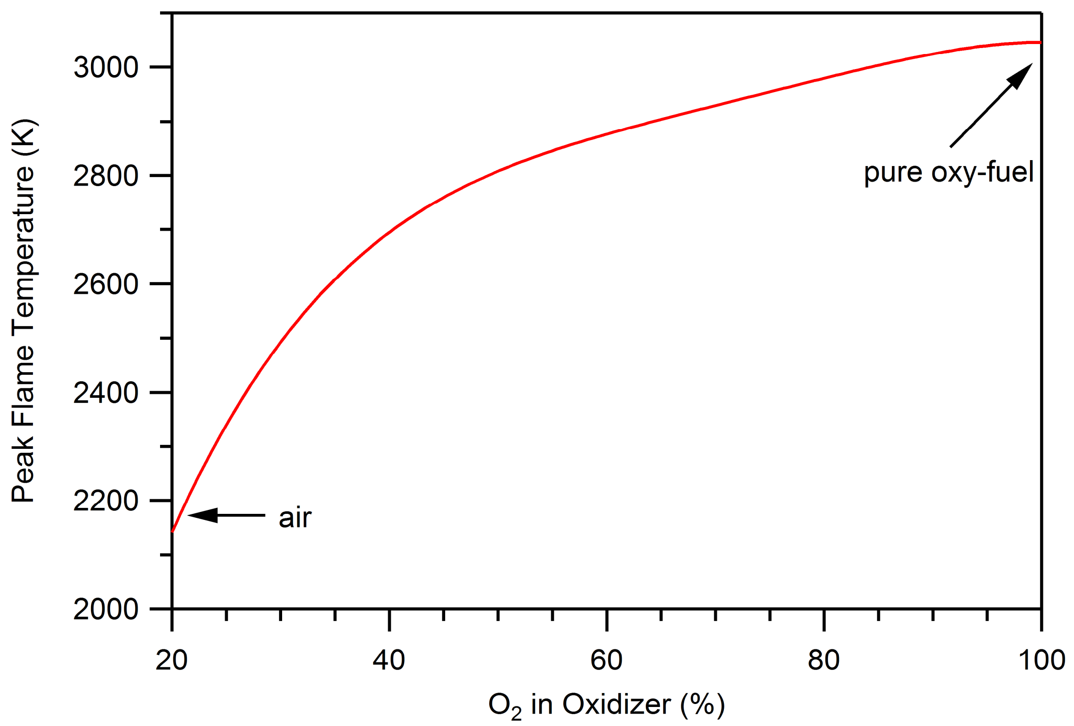
Beginning in mid-1980's, many glass melting furnaces and metal forging/shaping furnaces have been converted to oxy-fuel combustion

- **increases radiant heat transfer** (greater thermal efficiency of furnace)
  - higher flame temperature
  - (3.5x) higher concentration of radiant products (CO_2 and H_2O)
  - longer residence time in furnace
- **reduces emissions** (NOx, SOx, particulates)



steel reheat furnaces

Effect of O_2 Content on Flame Temperature (methane fuel, N_2 as diluent)



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Background

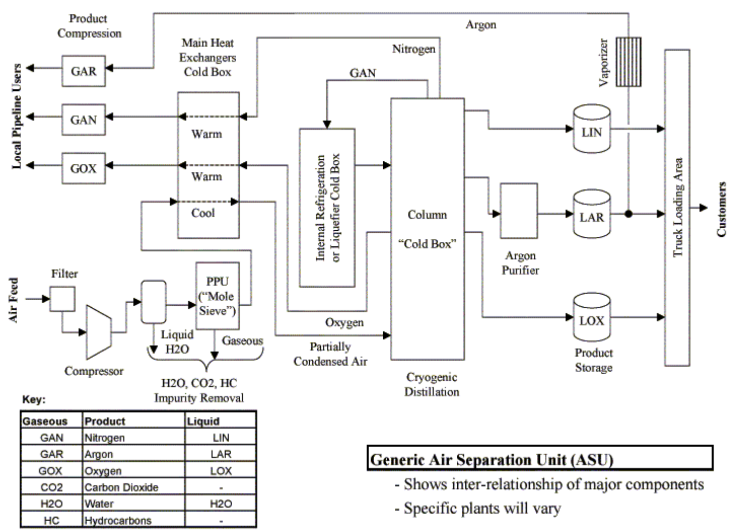
- Because O_2 is supplied (at cost), oxy-fuel furnaces operate just slightly fuel-lean (based on CO emissions): $\phi \approx 1$
- With premium on radiant heat transfer, soot formation in flame is desired (non-premixed combustion)

sooty flame in oxy-fuel glass melting furnace

O_2 Production

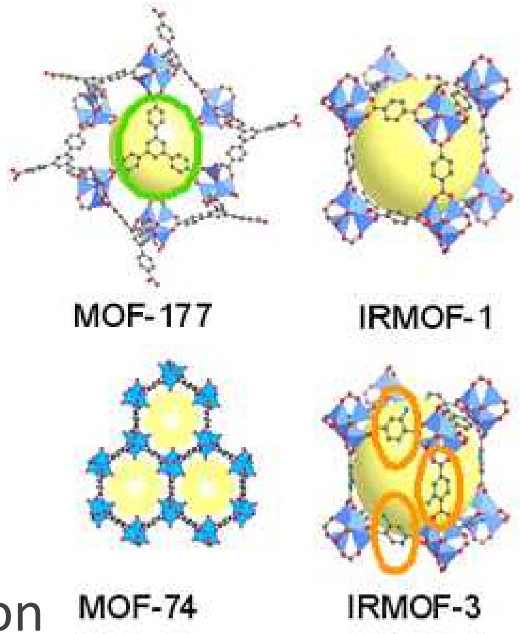
- To-date, oxy-fuel furnaces have used high-purity O_2 (95+%) from cryogenic air separation
- Cryogenic air separation has a high capital cost, limiting technology penetration to large facilities and providing small improvement in overall energy efficiency of entire system (though furnace efficiency can be dramatically increased)

Dual cryogenic separation towers, Callide Oxyfuel Demo Project, Australia



New Trends in O_2 Production

- Metal-organic frameworks (**MOFs**) is a new class of engineered materials that features very high surface areas for gas adsorption
- MOFs give possibility of low-cost air separation by attracting (electrophilic) O_2 to the metal coordination sites
- Sandia/NM, and others, are investigating the generation of new MOFs for efficient air separation



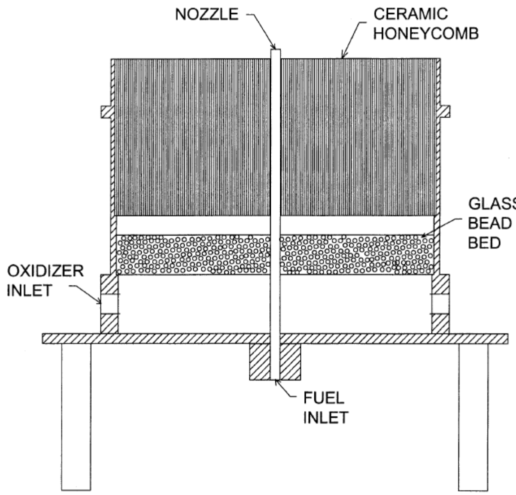
- Commercial air separation by MOFs will likely occur using pressure swing adsorption (PSA), whose economics will likely limit O_2 purity that can be generated
- Ion transport membrane (ITM) air separation also produces impure O_2

Motivation for This Investigation

Effect of O_2 purity on soot formation and total radiation from non-premixed turbulent natural gas oxy-fuel flames is not well understood

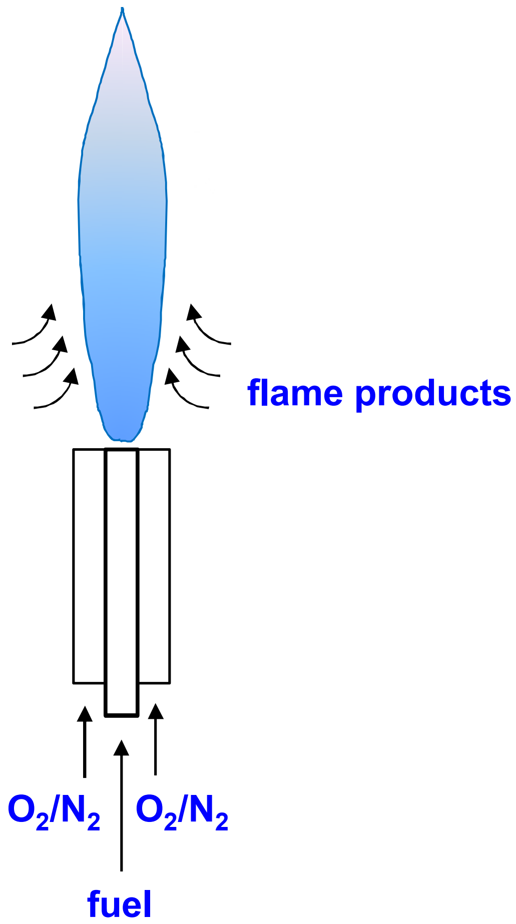
Previous Work

- Steve Turns' group at Penn State Univ. investigated turbulent propane and natural gas fuel jets in nearly quiescent laminar oxidizer co-flow (CST 2002)
  - fuel jet Reynolds numbers of 5000 to 15000
  - 21% to 100% O_2
  - wide-angle radiant emission
  - line-of-sight soot concentration (laser extinction)
  - NOx and CO emissions



- Soot concentrations peaked for 30-40% O_2 for Re =15000 and for 75% O_2 for Re = 5000, then decreased rapidly for higher O_2 purity
- Total flame radiation steadily increased with increased O_2 purity (as also found by Baukal and Gebhart, 1997, for non-sooting flames)
- **The PSU study did not address practical considerations of turbulent oxidizer flow, or only providing a stoichiometric amount**

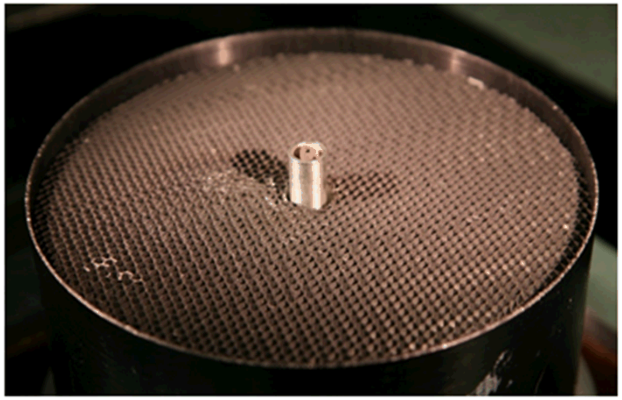
Practical Oxy-fuel Jet Flame Configuration

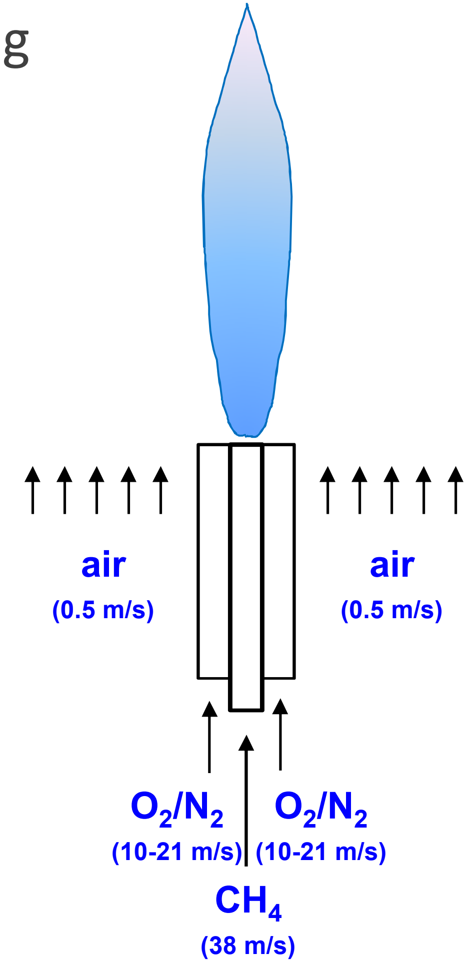


- turbulent mixing between central fuel jet and surrounding oxidizer jet
- turbulent mixing of oxidizer jet and surrounding stagnant (O_2-free) flame products
- **combined effects of O_2 concentration and turbulent mixing**, since lower O_2 concentration in oxidizer is, by necessity, associated with higher oxidizer flow (to maintain stoichiometric conditions)

Experimental Methods (Current Study)

- fuel/oxidizer coannular geometry with a surrounding low velocity coflow of air (or N_2)
  - stainless steel fuel tube (3.3 mm ID)
  - stainless steel oxidizer tube (10.2 mm ID)
- two flame series studied, in which either oxidizer composition or flow rate (for 100% O_2) were varied





Experimental Matrix

Flame Series	CH_4			Oxidizer				ϕ †	T_{ad} ‡ (K)	Shield gas flow
	slpm *	m/s	Re	% O_2 ¥	slpm	m/s	Re			
Constant Mixing	20.0	38.0	7680	38	40.0	10.4	3660	2.67	1467	air
	↓	↓	↓	50	↓	↓	↓	2.0	2240	↓
	↓	↓	↓	75	↓	↓	↓	1.33	2949	↓
	↓	↓	↓	100	↓	↓	↓	1.0	3054	↓
Variable Mixing	20.0	38.0	7680	100	40.0	10.4	3660	1.0	3054	air
	↓	↓	↓	↓	60.0	15.6	5490	0.67	2971	↓
	↓	↓	↓	↓	80.0	20.8	7320	0.5	2861	↓
	↓	↓	↓	↓	80.0	20.8	7320	0.5	2861	N_2

* standard conditions defined to be 298 K and 1 atm. pressure

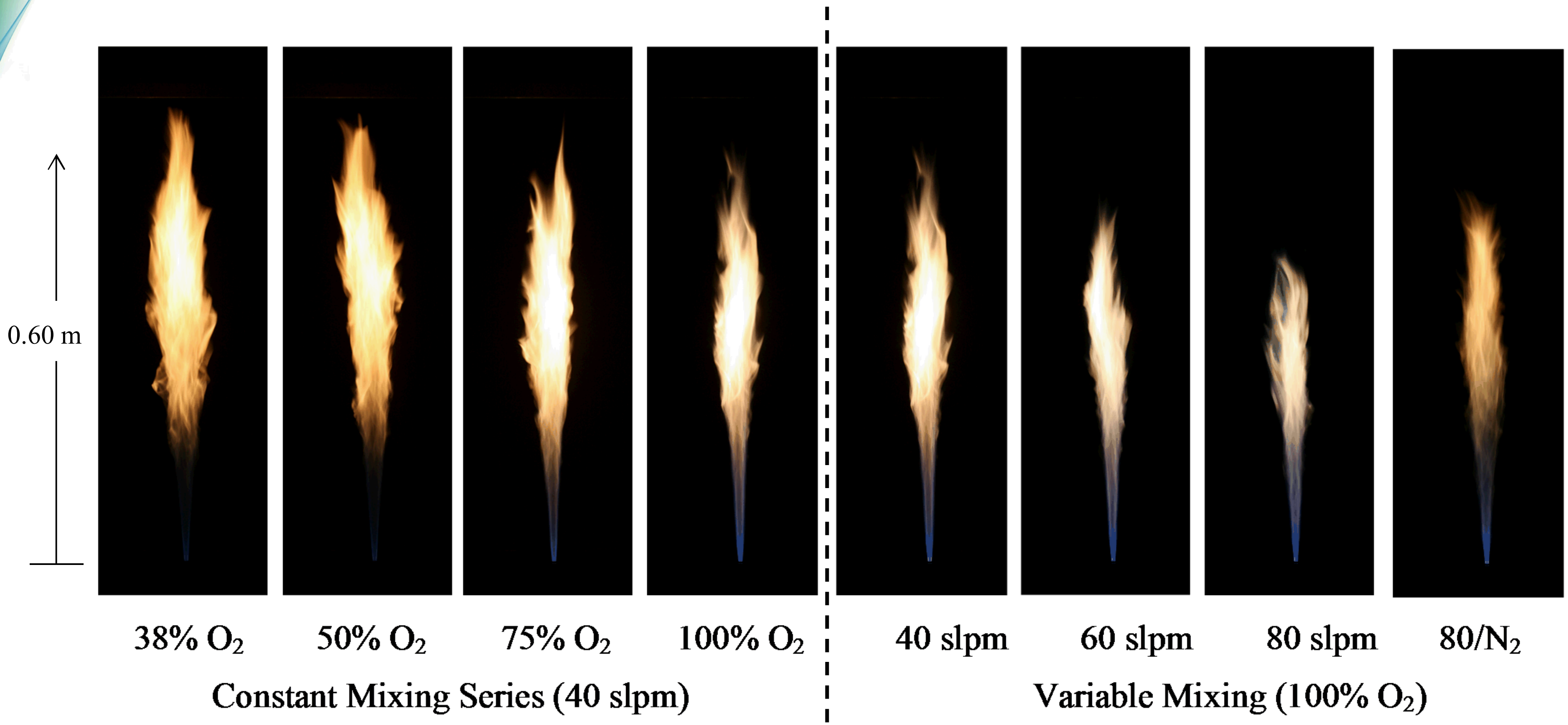
¥ balance gas of N_2

† stoichiometry of supplied gases

‡ adiabatic flame temperature of supplied gases (calculated using NASA cea code)

- all flames appeared to be attached
- heat release rate was 12.1 kW

Photographs of Flames

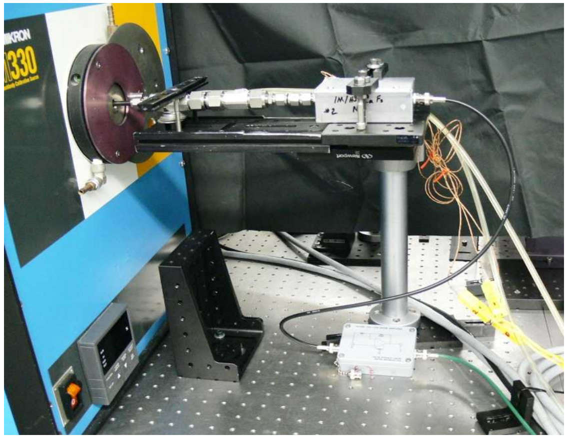


Measurements Performed

- Narrow-angle total radiant emission
- Planar Laser-Induced Incandescence (PLII) – soot concentration fields
- Planar soot two-color pyrometry – soot T (data is being analyzed)

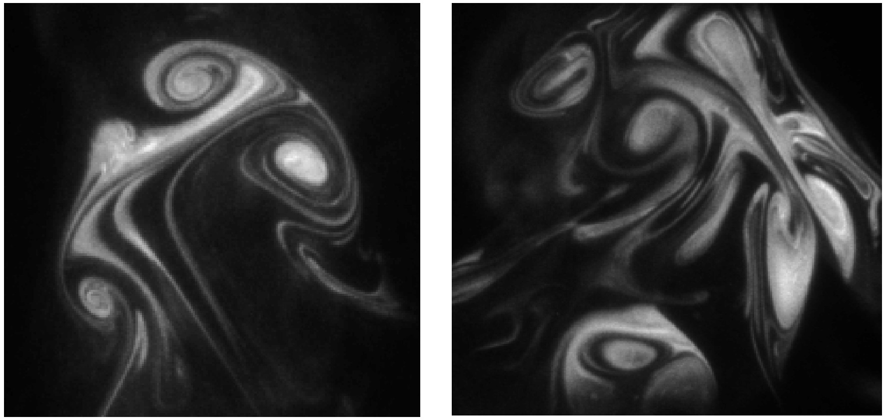
Radiometry Measurements

- thin-film thermopile with a CaF_2 window
- sensitive from 0.13–11 μm
- 32 ms response time
- black-anodized, 100 mm long steel tube (3 mm ID), restricted detection solid angle (Ω) to 1.61×10^{-3} sr (cone 9 mm wide at flame)
- 10 sec time records recorded at 1000 samples/s
- calibrated with a blackbody



PLII Measurements

- 1064 nm laser sheet excitation (37 mm high, 52 mJ/pulse, 0.47 J/cm^2)
- LII detection through 600 nm short pass filter onto 512 x 512 Gen III PI-MAX ICCD with 100 ns prompt detection
- 2000-image averages with background correction
- f_v calibration against 3-pt Abel-inverted 532 nm extinction-derived soot volume fractions on laminar CH_4 flame, using Bouguer law with K_a = 6.7 (Williams et al., IJHMT 2007)



single-shot LII images

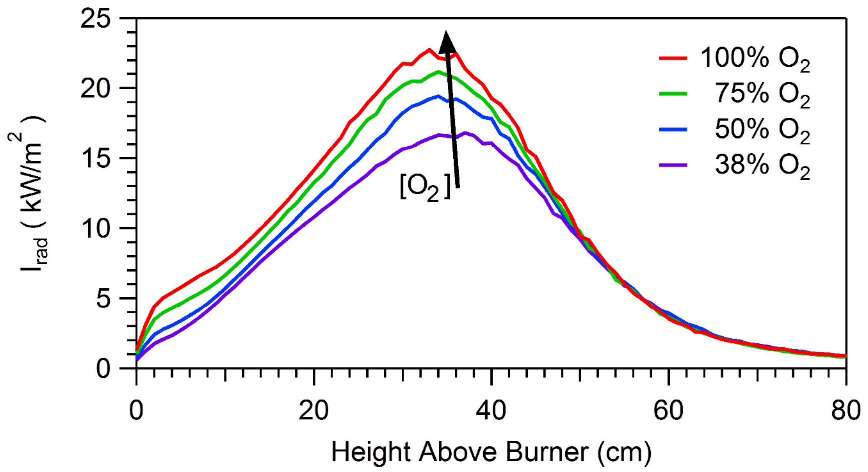
$$I/I_0 = exp^{-\int \frac{K_a}{\lambda} f_v dx}$$

Two-Color Pyrometry Imaging

- 50 mm beamsplitter with dual ICCD detection at 550 nm and 675 nm (20 nm bandwidth) – aligned to optical pattern at flame location
- 25 ms gate width; 400 image pairs at each location
- blackbody calibration, with high-temperature extrapolation verified with calibrated tungsten lamp (2919 K)
- assume $1/\lambda$ spectral variation of soot emissivity from 550 to 675 nm
  - using $1/\lambda^{1.39}$ (Hottel and Broughton) assumption would give soot temperatures 60-90 K lower

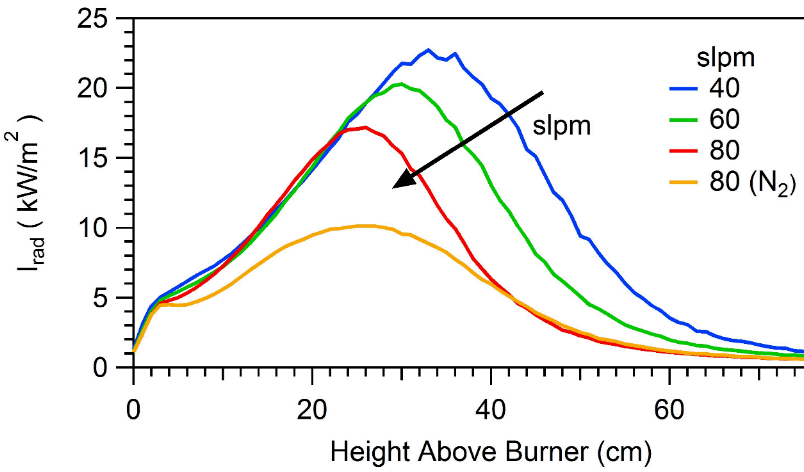
Experimental Results: Thermal Radiation (Centerline)

Effect of O_2 Purity



- For constant mixing, increasing $[O_2]$ results in somewhat enhanced radiation (7% lower for 75% O_2, 14% lower for 50% O_2)
- Radiant flame heights are invariant

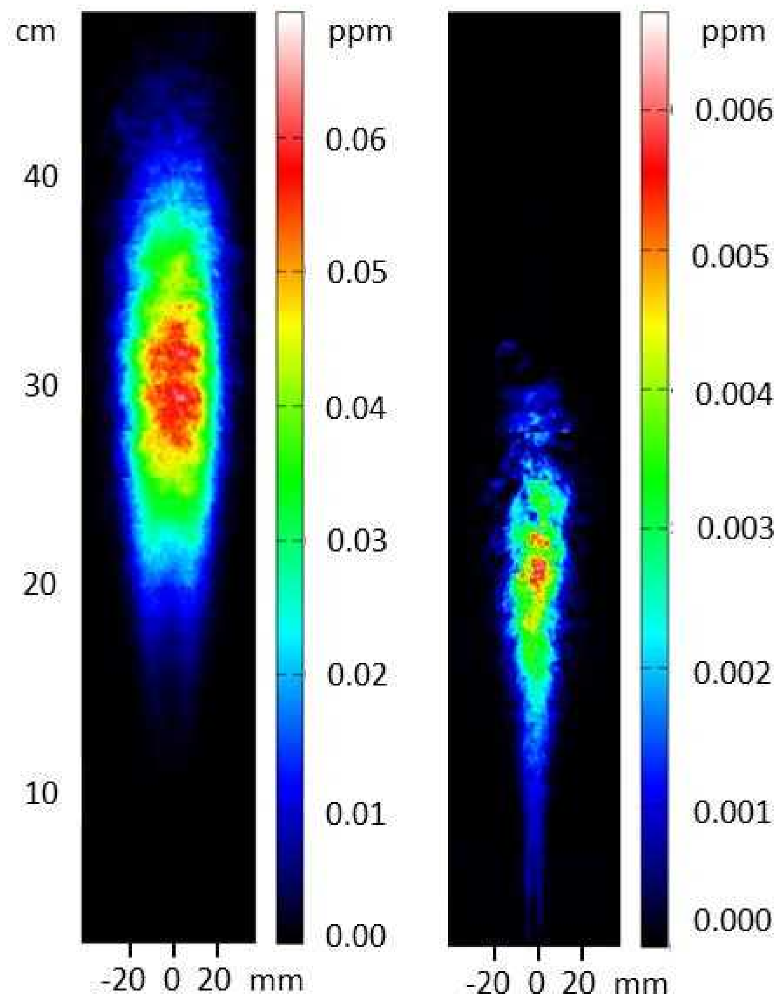
Effect of Turbulent Mixing



- For constant $[O_2]$, enhanced mixing results in much shorter flames and much lower radiation (20% lower for each 20 slpm)
- For strong mixing, N_2 ambient results in 25% lower radiation

PLII Results (Sample Mean f_v Fields)

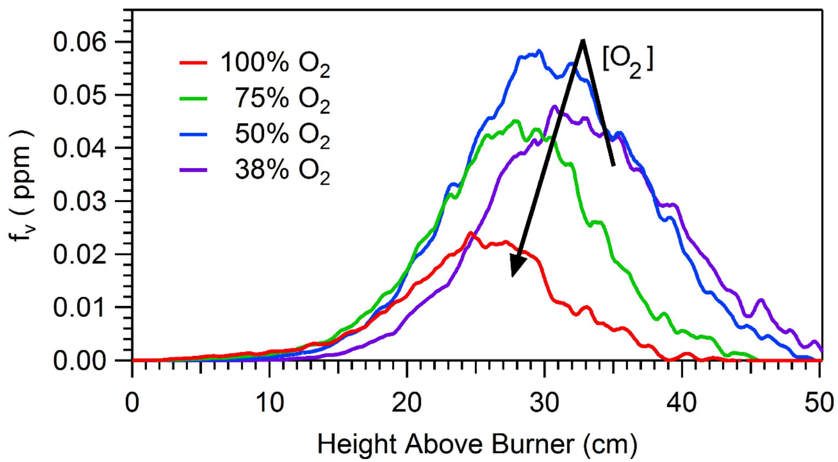
50% O_2
40 slpm
(most sooty flame)



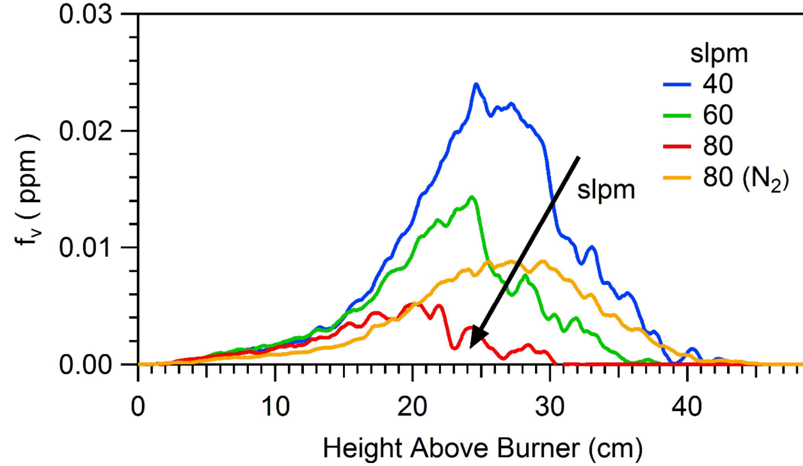
100% O_2
80 slpm
(least sooty flame)

Mean LII Results (Centerline)

Effect of O_2 Purity



Effect of Turbulent Mixing

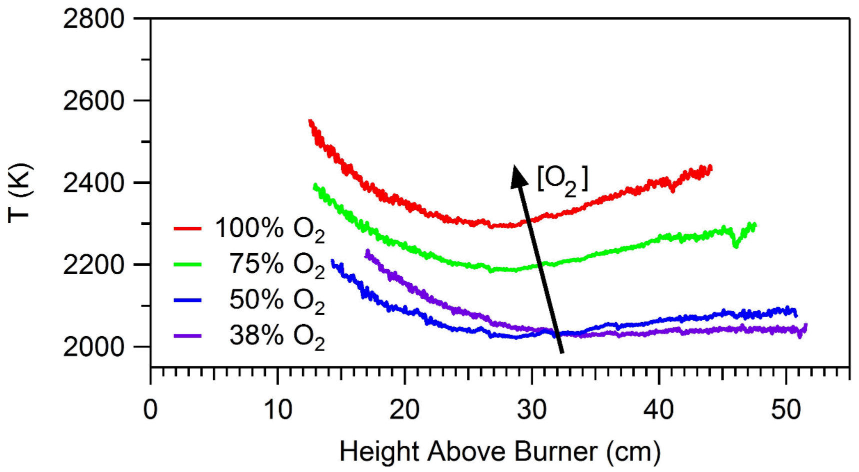


- For constant mixing, soot f_v peaks for ~ 50% O_2, then decreases rapidly as $[O_2]$ increases further

- For constant $[O_2]$, enhanced mixing results in much lower soot production
- For strong mixing, N_2 ambient results in enhanced, delayed soot production

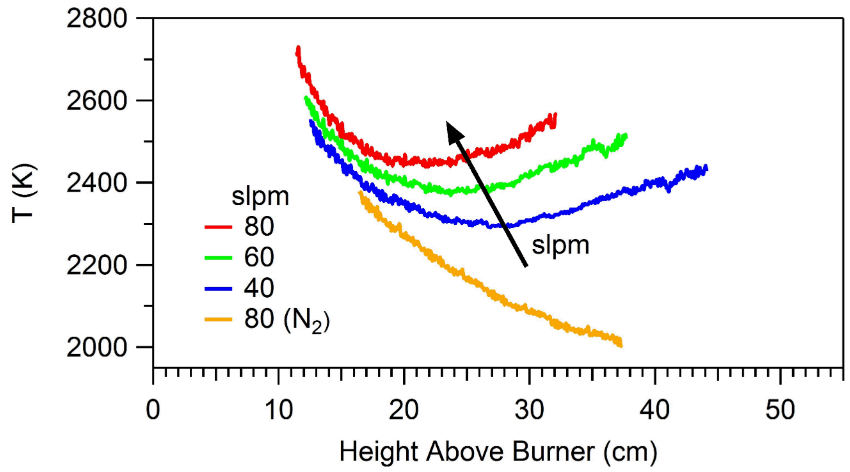
Measured Mean Soot Temperatures (Centerline)

Effect of O_2 Purity



- soot T asymptotes for 38% and 50% O_2 consistent with CH_4/air
- higher O_2 content gives hotter flame (also, has greater stoich. of supplied oxygen)

Effect of Turbulent Mixing



- tailing of T with N_2 ambient suggests continual dilution of supplied O_2 with ambient
- higher O_2 flow yields higher flame T (less dilution by ambient)

Interpretation of Results

- Soot T results demonstrate that entrainment of ambient gas into upper regions of flame (where soot forms) is important (also true of actual oxyfuel furnace flames, but ambient T/composition is different)
- As [O_2] decreases from 100% to 50% (in constant mixing flames), soot formation increases by factor of 3, tempering the reduction in thermal radiation to only 14%
  - Baukal and Gebhart found 200% variation in thermal radiation for [O_2] between 100% and 35% for non-sooting flames
- Increasing turbulent mixing intensity in pure oxyfuel flames has a marked influence on flame height, thermal radiation, and soot formation (all decreasing with mixing intensity), despite soot temperatures increasing
- Having a chemically inert ambient has a strong effect on thermal radiation (decreasing it) and soot production (increasing it)
  - minimizing the mixing rates between the 3 streams (fuel, oxidizer, ambient) is clearly desirable to optimize soot formation and radiation

Summary

- Recent developments in materials science point to potential low-cost methods for air separation, but yield impure O_2
- Varying the oxygen concentration in commercial oxyfuel applications necessarily impacts the oxidizer flow rate and turbulent mixing field (because ϕ constrained to be 1)
- We measured soot concentration, soot temperature, and radiation in two series of oxyfuel flames in which mixing intensity and $[O_2]$ were varied, separately
- Measurements show soot formation increases with decreasing purity of O_2 (at least down to 50% O_2), tempering the trend of lower flame radiation
- Turbulent mixing rates have large impact on soot formation, temperature, and flame radiation
- Minimizing the mixing rates between the 3 streams (fuel, oxidizer, ambient) is desirable to optimize soot formation and radiation

Acknowledgment

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Questions?