Experimental and Modeling Investigations of Ignition and Char Combustion during Oxy-Fuel Combustion of Pulverized Coal

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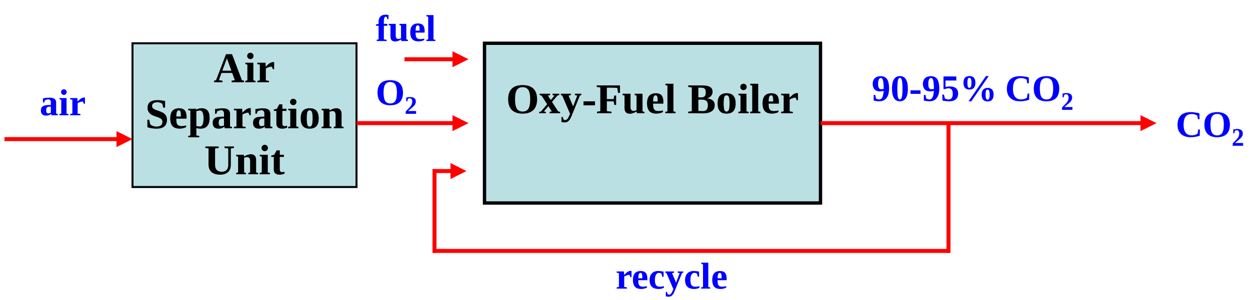
Oxy-Fuel Combustion

- One of the more promising options for carbon capture and sequestration (CCS) when using coal for power production:



- can be retrofitted to existing boilers
- modest modification of existing technology
- concurrent emissions reductions

- 30 MW demonstration project operational in Germany; others planned/under construction in Australia, U.S., Spain, Scotland
- Integration of membrane oxygen production and reduction of flue gas recycle promise efficiency improvements

Schwarze Pumpe pilot plant

Oxy-Fuel Combustion: What's Different?

- **Elimination of nitrogen diluent and its partial replacement with recycled CO_2 results in**
  - ✓ lower gas velocity
  - ✓ more concentrated product gases in the boiler
  - ✓ significant differences in gas transport properties
  - ✓ radiantly active gas medium (IR absorption and emission)
- **Improved control over flame temperature, flame stabilization, and carbon burnout, by controlling O_2 level in different flow streams**
  - ✓ primary, secondary and tertiary air streams replaced with different mixtures of O_2/recycled flue gas
  - ✓ concentrated O_2 lances can be used to promote flame stabilization

Motivation for Bench-Scale Studies of Oxy-Fuel Combustion

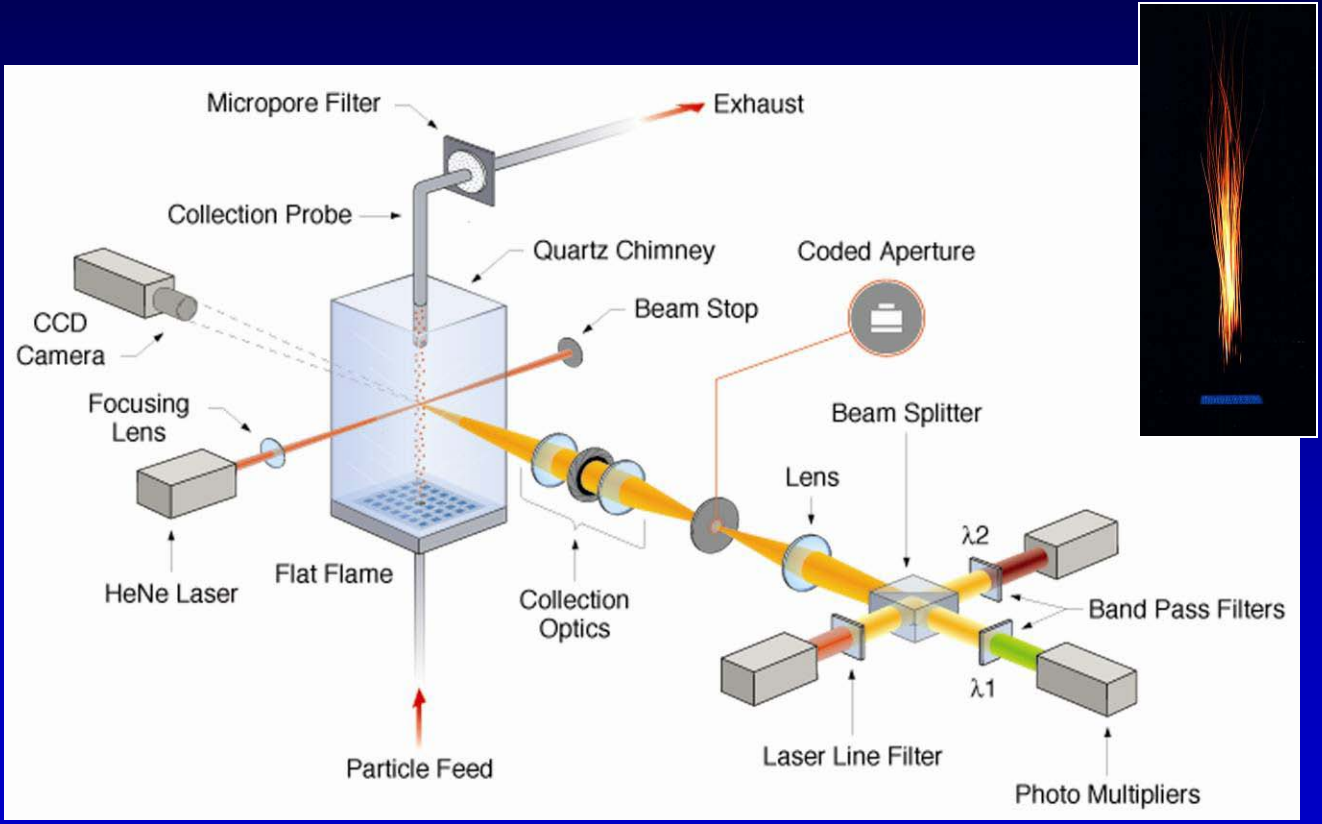
- **Better control of important variables:**

  temperature
  velocity
  local gas mixture ([O_2], [CO_2], [H_2O])
  particle size
  particle loading
  coal vs. char
  residence time
  etc.

  – allowing clear identification of governing phenomena

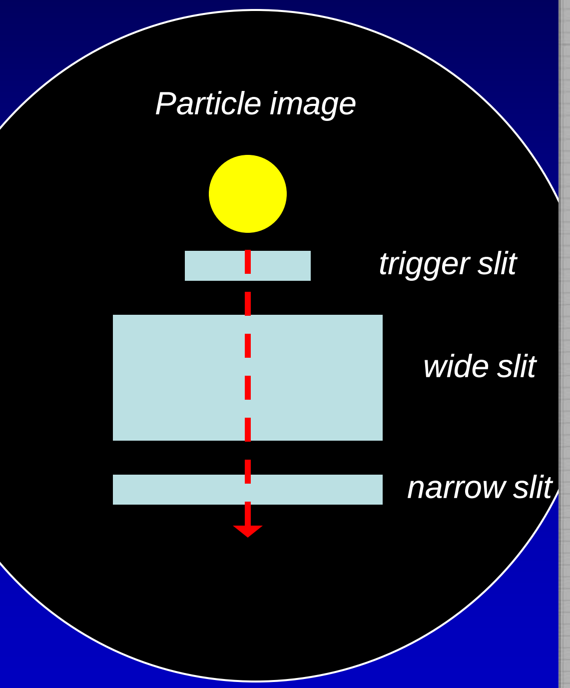
- **Ability to apply advanced diagnostics**
- **Ability to quantify rate parameters for use in CFD models**

Experimental Setup: Combustion-Driven Optical Entrained Flow Reactor

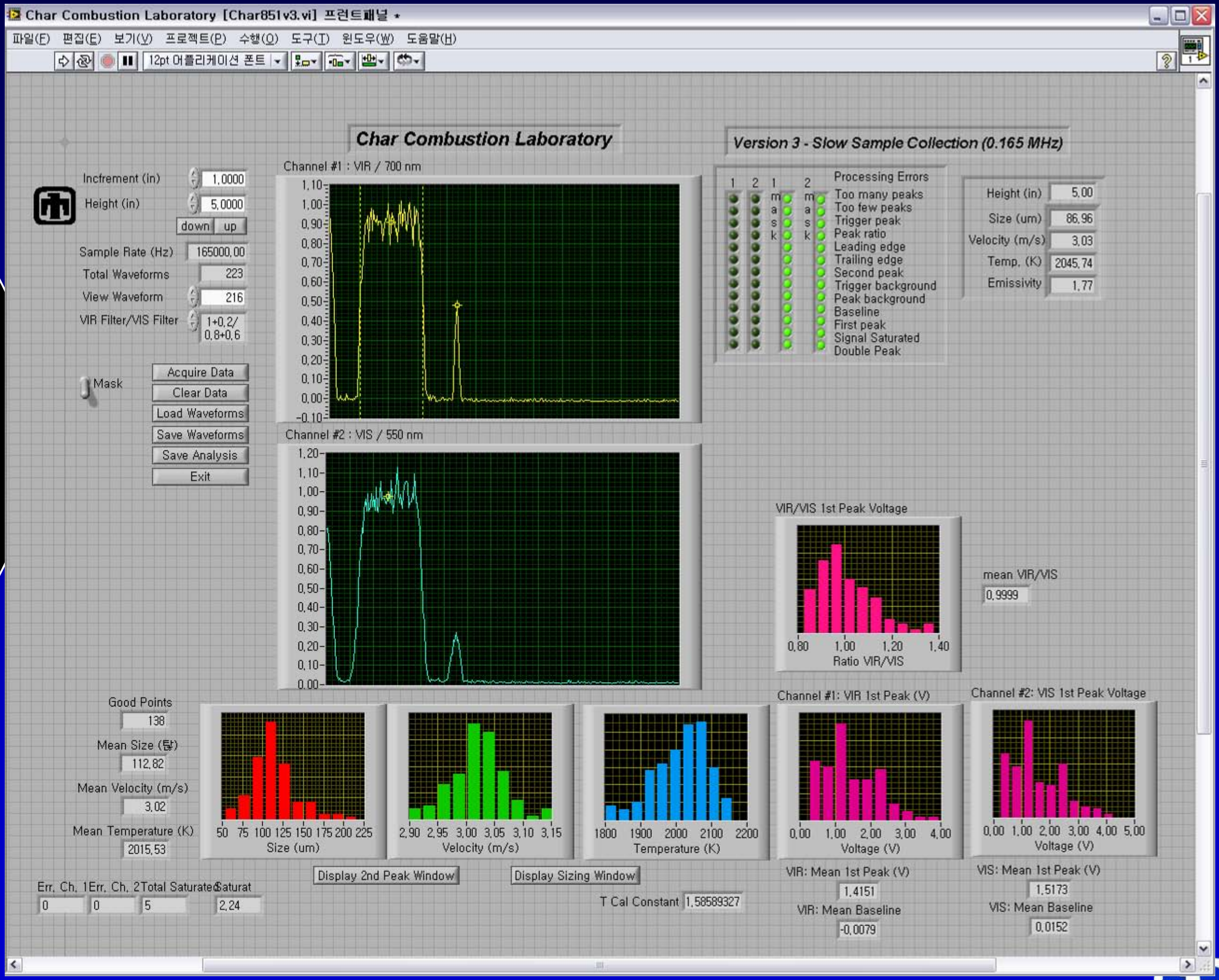


- 1 atm
- compact, diffusion-flamelet burner
- coal or char particles introduced along centerline
- quartz chimney
- coded-aperture, 2-color pyrometry diagnostic for char size, T, and velocity
- laser-triggered CCD/ICCD for particle imaging

Particle-Sizing Pyrometry in the Laminar Entrained Flow Reactor

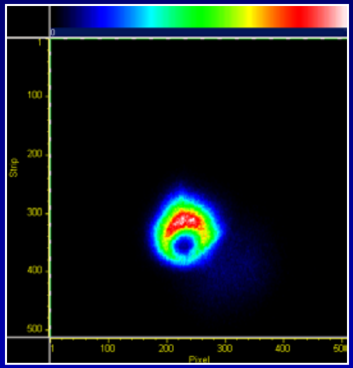


schematic of coded aperture



Previous Oxy-Combustion Studies in SNL LEFR

- **Ignition of isolated particles**
  - Pittsburgh hvb coal and Black Thunder subbit coal (75-106 μm)
  - 12 – 36% O_2 in N_2 and in CO_2 at 1250 K and 1700 K
  - measured time to ignition and duration of devolatilisation
  - soot and char particle temperatures measured
- **Combustion of isolated coal char particles**
  - full spectrum of coals, ranging from Beulah lignite through Black Thunder subbit coal to Pittsburgh hvb coal and anthracite
  - 75-106 μm particles burning in 12 – 36% O_2 in N_2 and in CO_2 at 1250 K and 1700 K
  - measured char particle temperatures and sizes



Thermal image of soot layer surrounding devolatilising Pittsburgh coal particle

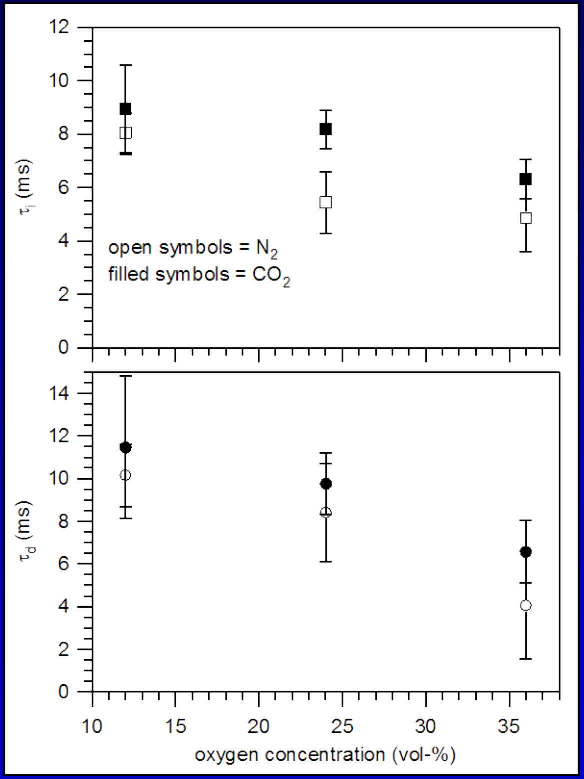
Conclusions from Study of Isolated Particle Ignition

- **ignition time and devolatilisation rate are sensitive to local oxygen concentration**
- **CO_2 retards particle ignition (slightly) and decreases devolatilisation rate (slightly)**
- **results consistent with theories of adiabatic thermal explosion and droplet combustion (CO_2 c_v and D_{fuel} effects)**

$$\tau_i = \frac{c_v \left(T_0^2 / T_a \right)}{q_c Y_{F,0} A \exp\left(-T_a / T_0 \right)}$$

$$\dot{m} = \left(4\pi r_p \right) \left(\rho_p D_{fuel} \right) \ln\left(1 + B \right)$$

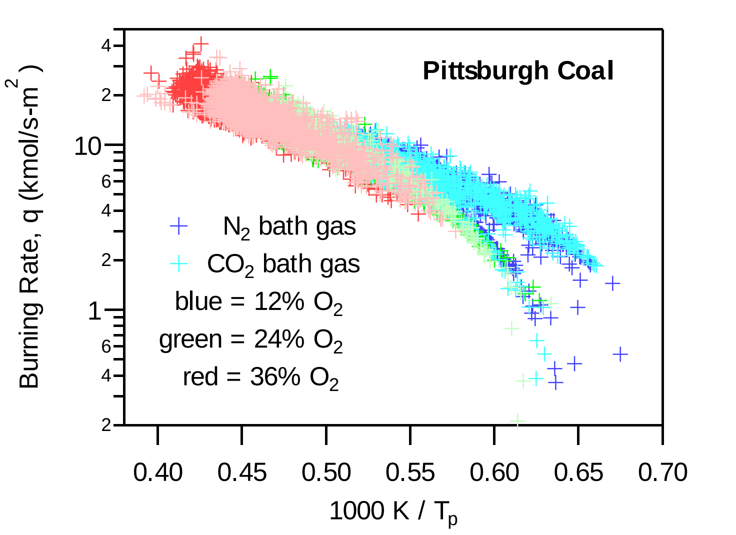
$$B = \left[c_{p,\,fuel} \left(T_\infty - T_p \right) + \left(Y_{o,\infty} / OF \right) h_c \right] \Big/ h_v$$



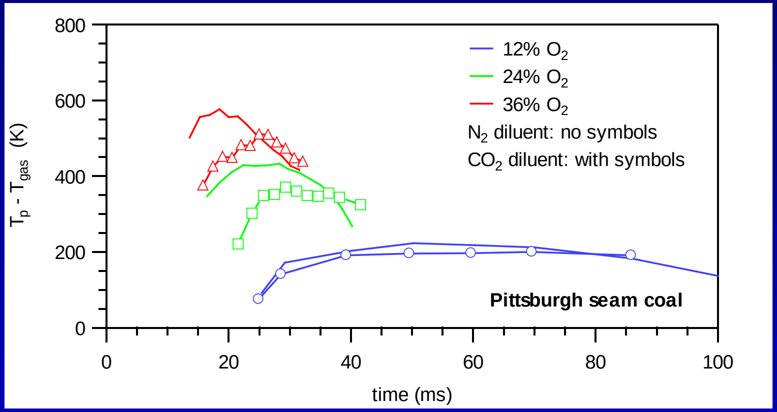
Measured Pittsburgh coal particle ignition times (top) and duration of devolatilisation (btm)

Conclusions from Studies of Isolated Char Particle Combustion

- **CO_2 reduces char particle combustion temperature, in amount consistent with reduction in O_2 diffusion through boundary layer**
- **apparent single-film char kinetic rates unaffected by CO_2**



Arrhenius plot of measured Pittsburgh coal char particle surface specific burning rates in N_2 and CO_2 atmospheres



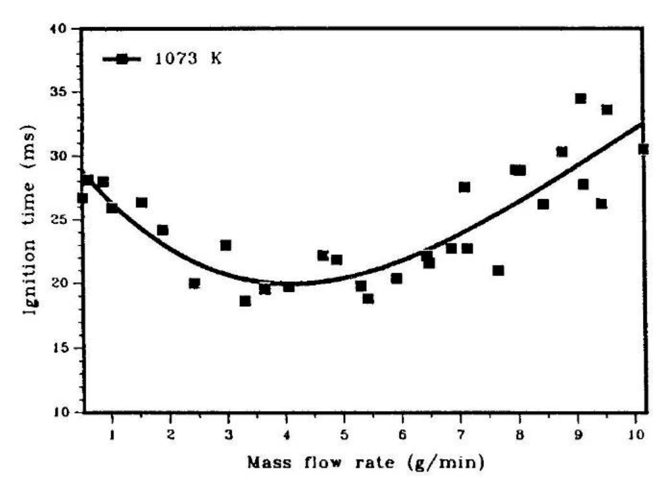
Mean particle rise of Pittsburgh coal char particles in N_2 and CO_2 atmospheres

Current Oxy-Combustion Studies at SNL

- Ignition of high concentration of pc particles
- Evaluation of influence of boundary layer chemistry and CO_2 char gasification reaction on pc char combustion

Motivation for Coal Stream Ignition Study

- **Several studies have shown poorer ignition quality in oxy-fuel flames (depending on O_2 level, type of coal, type of burner, etc.)**
- **Gas stream momentum differences and inherent ignitability differences (relative to air-fired) complicate understanding of flame-holding in oxy-fuel combustion**
- **Very limited data available (even for conventional conditions) on coal stream ignition in laminar flow, for development/validation of CFD models**
  - Ruiz, Annamalai, and Dahdah, HTD 1990
  - hv bit coal (Pee Wee), 53-75 μm
  - 9 vol-% O_2
  - ignition point via thermal image on camera

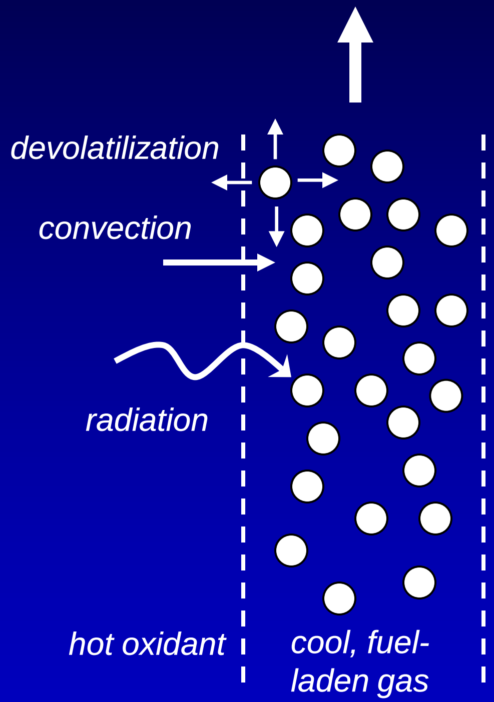


Influence of Particle Loading on Ignition of Pulverized Coal Particles

- **Limited experimental and modeling studies of effect of particle loading on pc ignition**
- **Characterize particle loading with Group number**

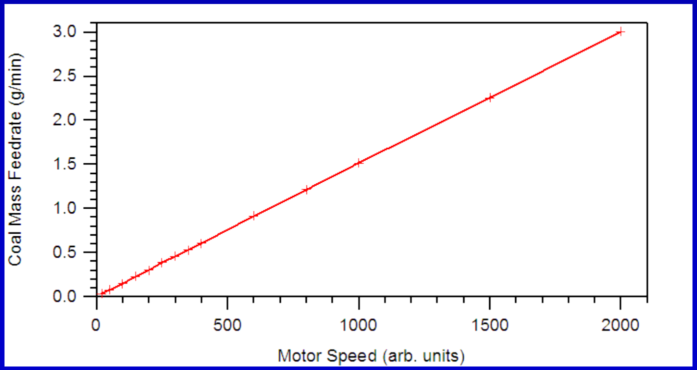
  $$G \sim n_p * d_p * r_{cloud}$$

- **Competing effects as particle loading increases**
  - presence of merged volatiles clouds promotes mixing of volatiles with hot ambient (decreasing ignition delay)
  - at high particle loading, sheltered inner region of particles absorbs heat without yielding substantial volatiles (increasing ignition delay)
  - minimum in ignition delay as function of Group number



Coal Ignition Study

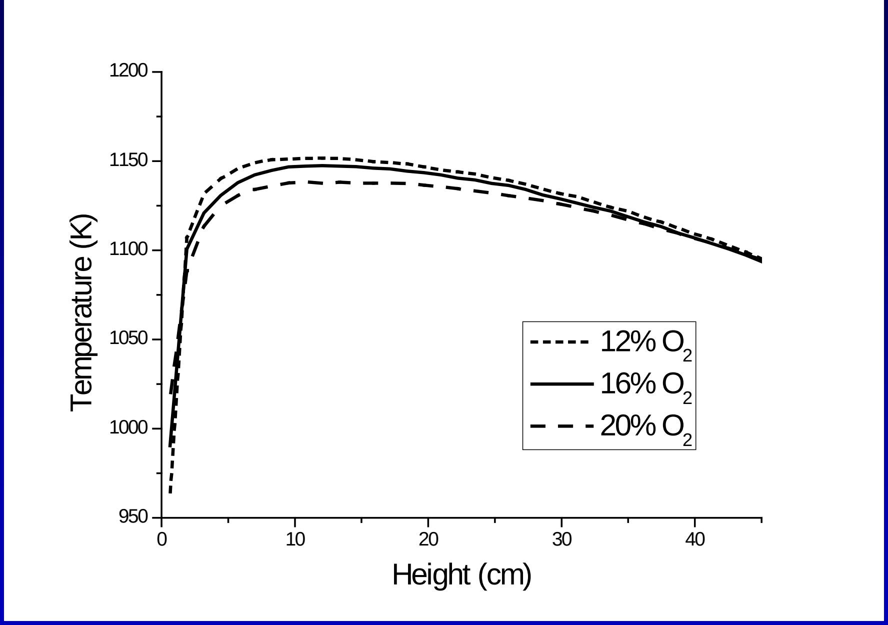
- **Enabled by installation of new coal particle feeder that produces steady coal flow rates up to 4 g/min through small diameter steel tubing**
  - design is modified version of concept developed in Prof. Sarofim's lab at MIT
  - feed rate determined by rate of displacement of coal-containing test tube
  - similar feeders in use at Univ. of Utah and U.S. EPA



Coal feed calibration plot

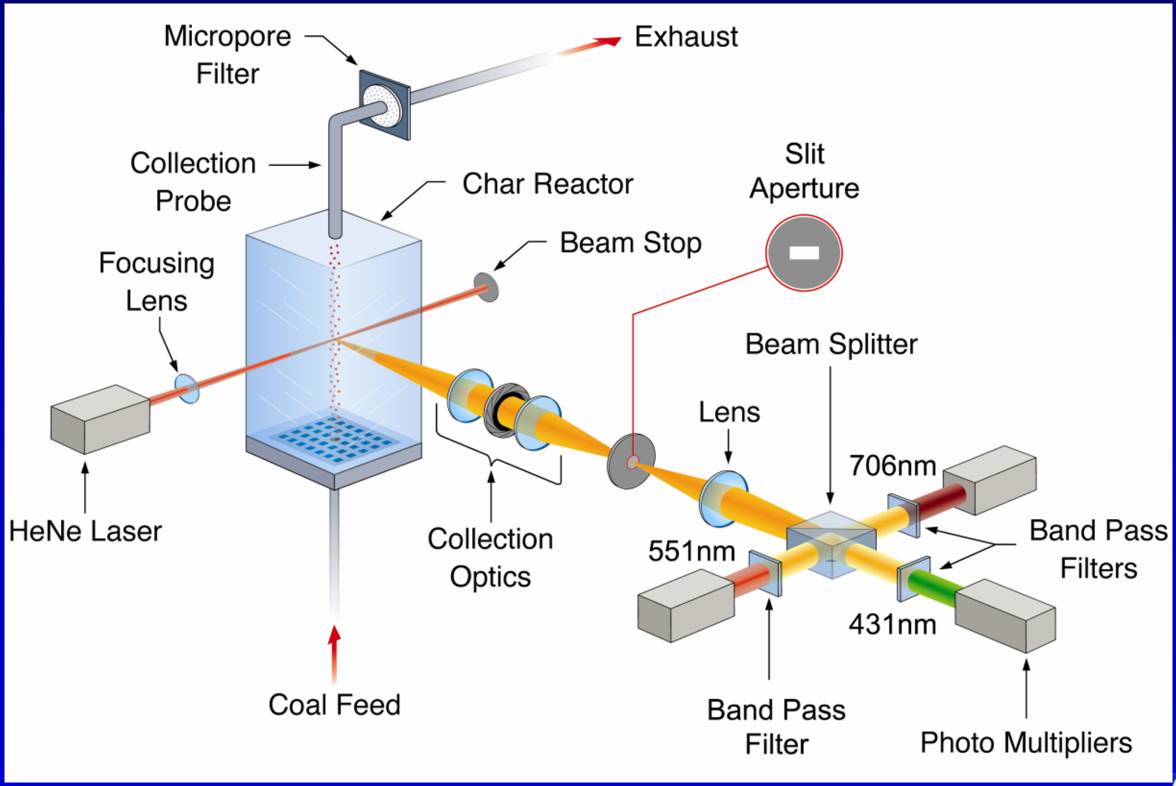
Photograph of pulverized coal feeder

Gas Temperature Profile



Optical Setup

- **Collect**
  - ✓time-lapse visible light emission
  - ✓2-color pyrometry of individual particles
  - ✓CH* emission



Photographs of Coal Flow Ignition

12 vol-% O_2 in N_2 bulk gas

Pittsburgh coal

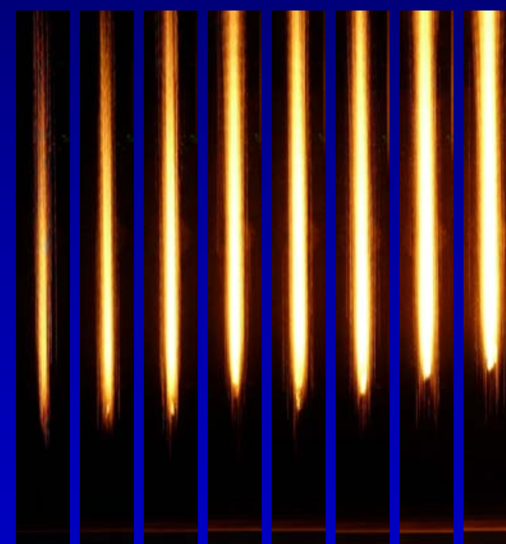
flow

burner surface

1 2 3 4 5 6 8 10

feedrate (x 0.06 g/min)

Black Thunder coal

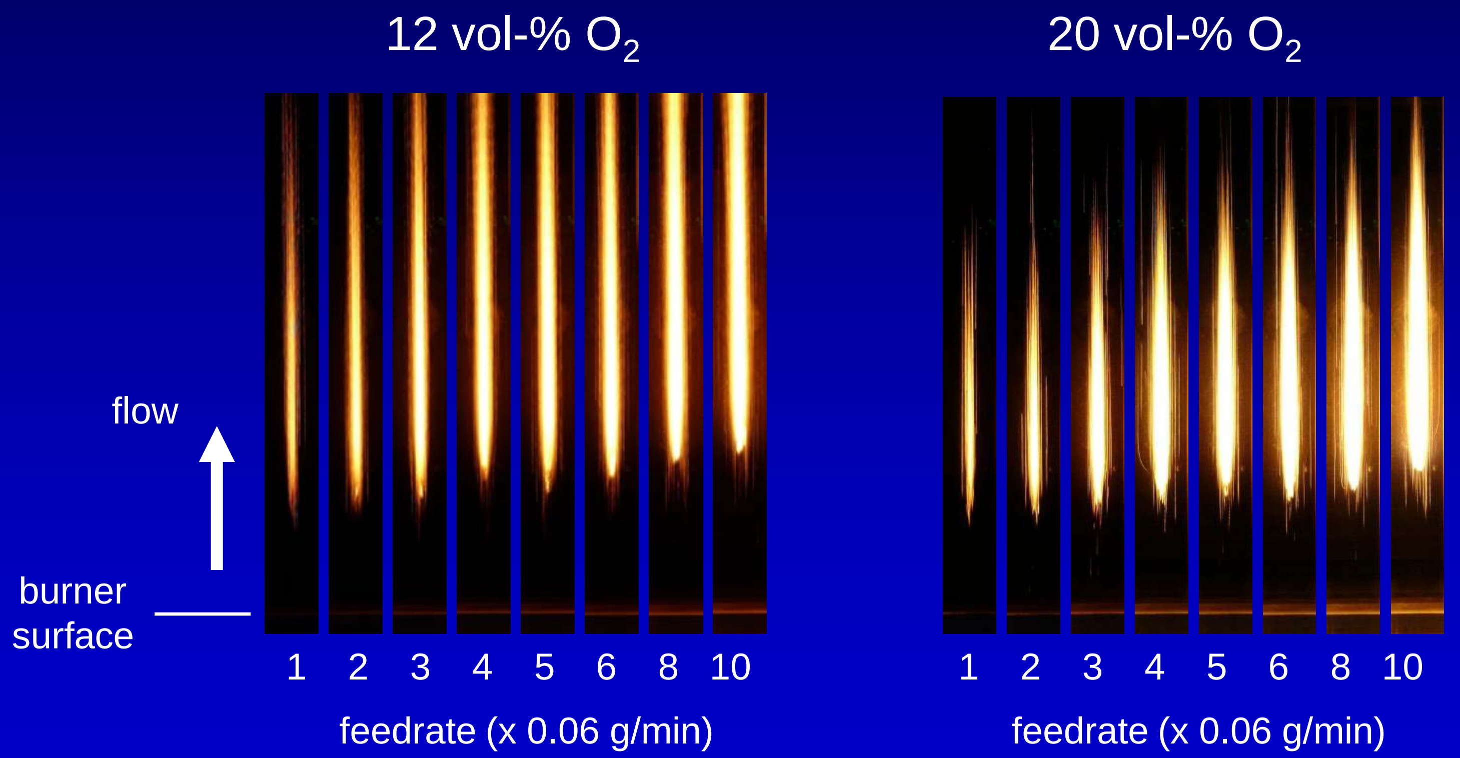


1 2 3 4 5 6 8 10

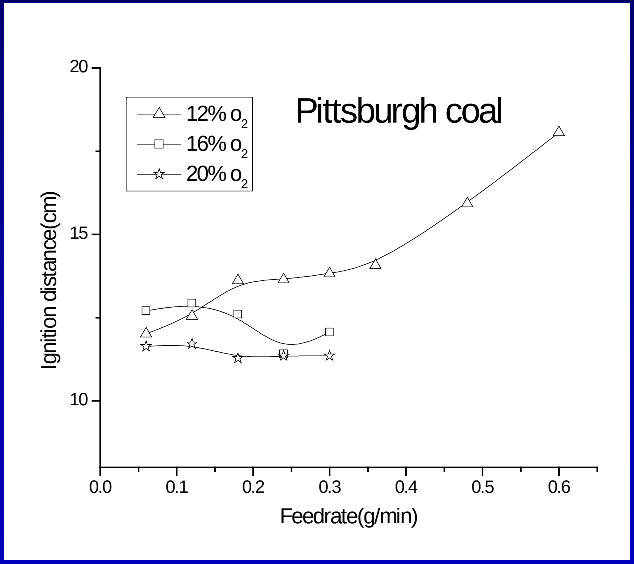
feedrate (x 0.06 g/min)

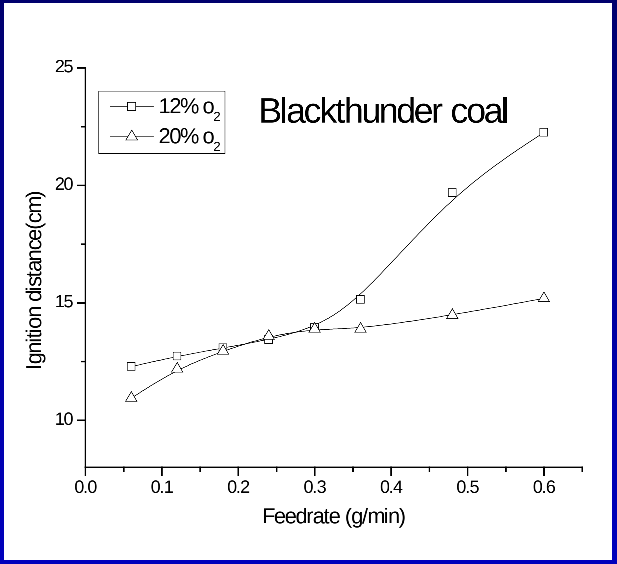
Photographs of Coal Flow Ignition

Black Thunder coal, N_2 bulk gas



Ignition Height Profiles



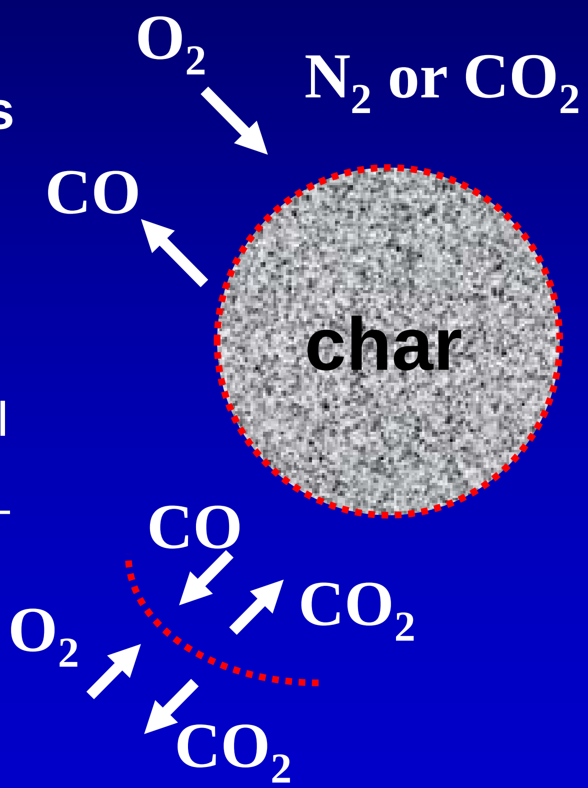


Influence of Boundary Layer Conversion of CO during Oxy-Fuel pc Char Combustion

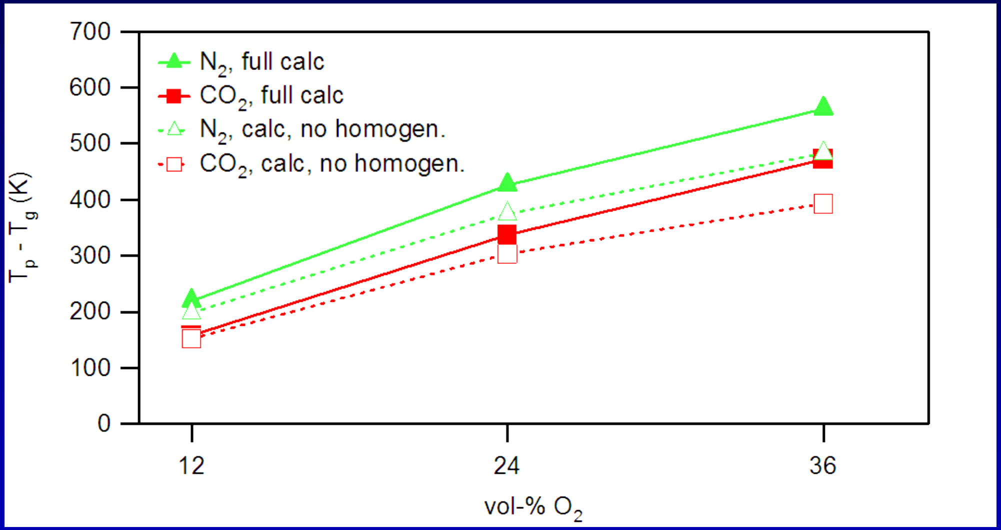
- **Analyze with SKIPPY code, written by Prof. Brian Haynes (Univ. Sydney)**
- **SKIPPY = Surface Kinetics in Porous Particles**
- **GRI-MECH 3.0 gas-phase kinetics; CHEMKIN II surface kinetic approach**
- **Our simulations:**
  - simple adsorption/desorption char oxidation model

	A (g/cm^2-s)	E (kcal/mol-s)
C_s + O_2 => CO + O_s	3.30E+15	40.0
O_s + 2C(b) => CO + C_s	1.00E+08	0.
C_s + O_2 => O_2_s + C(b)	9.50E+13	34.0
O_2_s + 2C(b) =>C_s + CO_2	1.00E+08	0.

  - typical char properties, experimental conditions

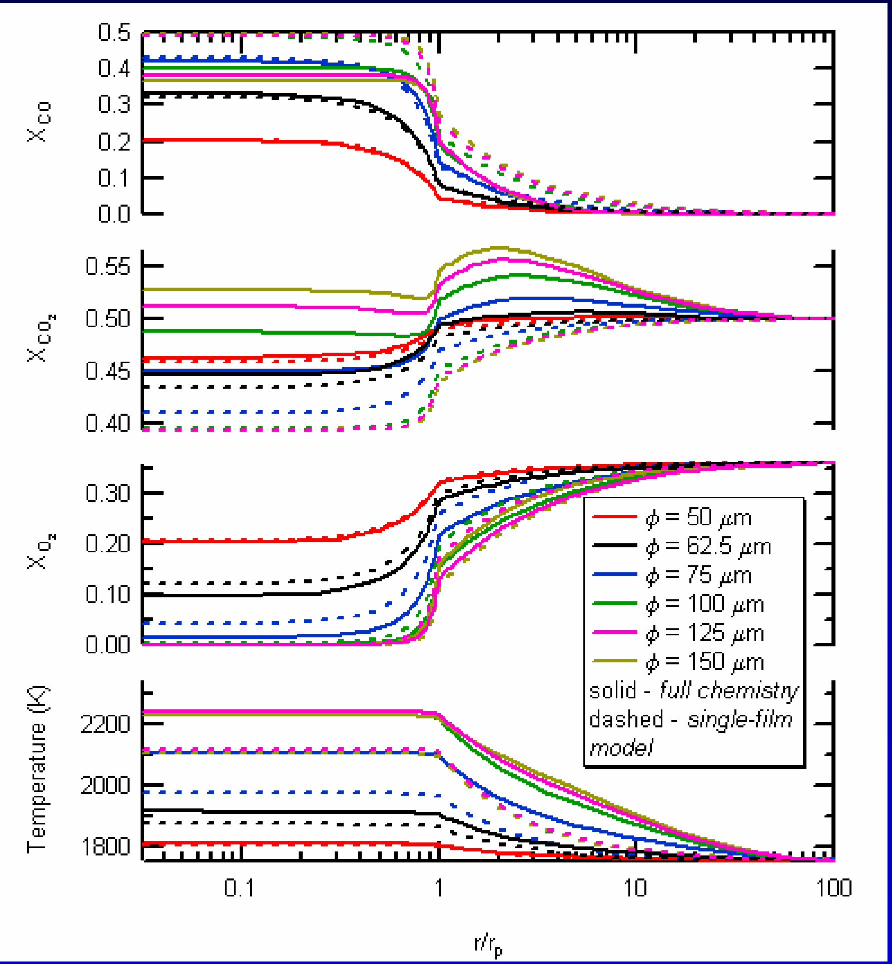


SKIPPY Results: Effect of Boundary Layer Reactions as Function of O_2



- **Boundary layer conversion becomes increasingly important for combustion in higher O_2 levels (greater boundary layer T)**
- **Some effect seen even for combustion in 12 vol-% O_2**
- **Calls into question general applicability of single-film model (in CFD codes and in deriving char kinetic rates)**

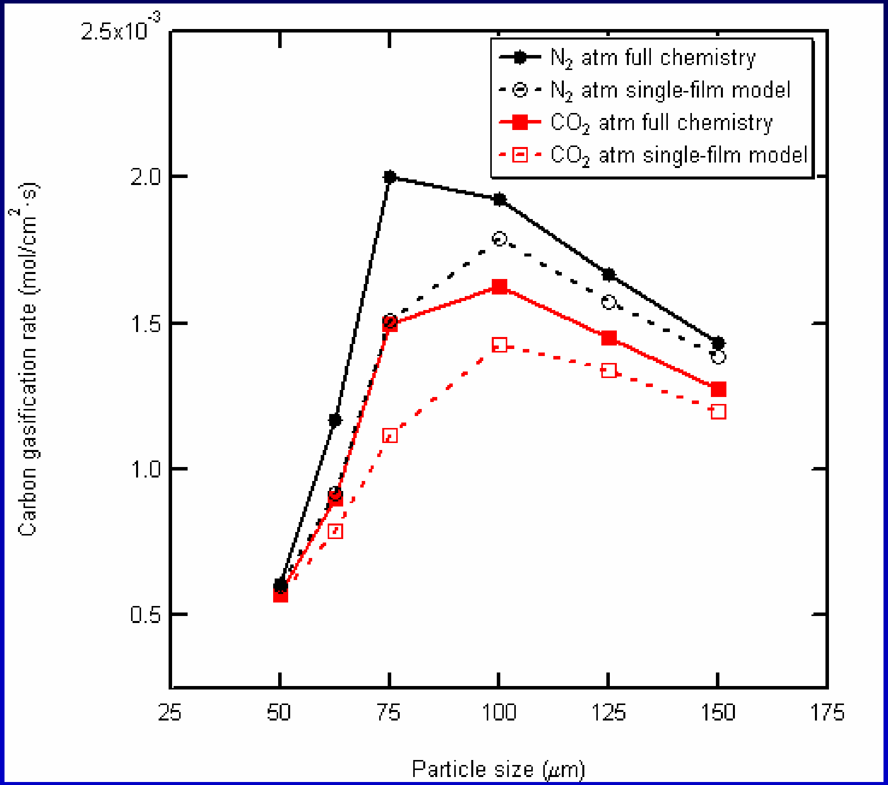
SKIPPY Results: Particle Size Effect of Boundary Layer Reactions



- Assumed gas composition
  36% O_2
  14% H_2O
  50% CO_2
- CO_2 is produced in boundary layer
- Single film model (heterogeneous chemistry only) **under-predicts particle temperatures** for sizes larger than 60 μm
- Particles larger than 100 μm are burning near the diffusion limit

full chemistry (solid lines); single-film model (dashed lines)

SKIPPY Results: Particle Size Effect of Boundary Layer Reactions



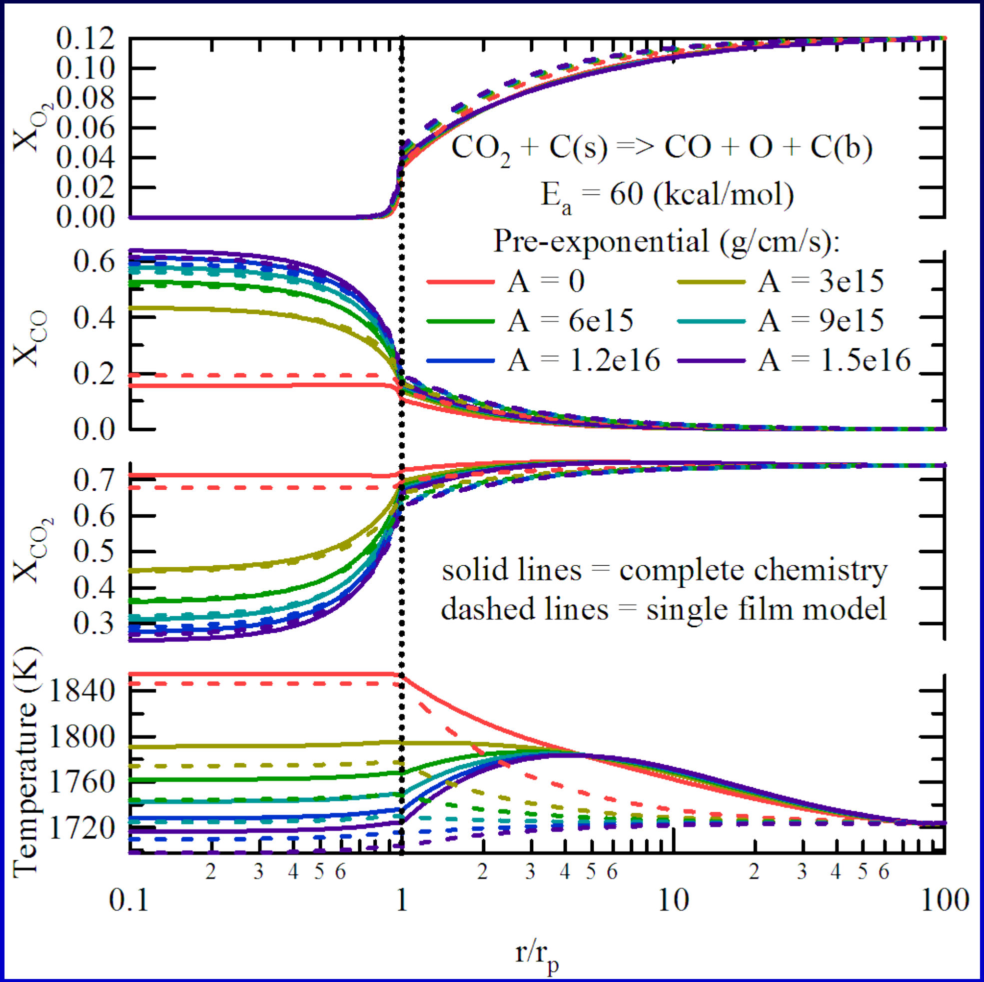
full chemistry (solid lines); single-film model (dashed lines)

- **Particle reaction rate peaks for size range around 70-80 μm**
  - Diffusion time scales for CO and O_2
  - convective, radiative heat transfer
  - effect of particle temperature on boundary layer conversion rates
- **Single-film burnout rate underpredicts actual rate**

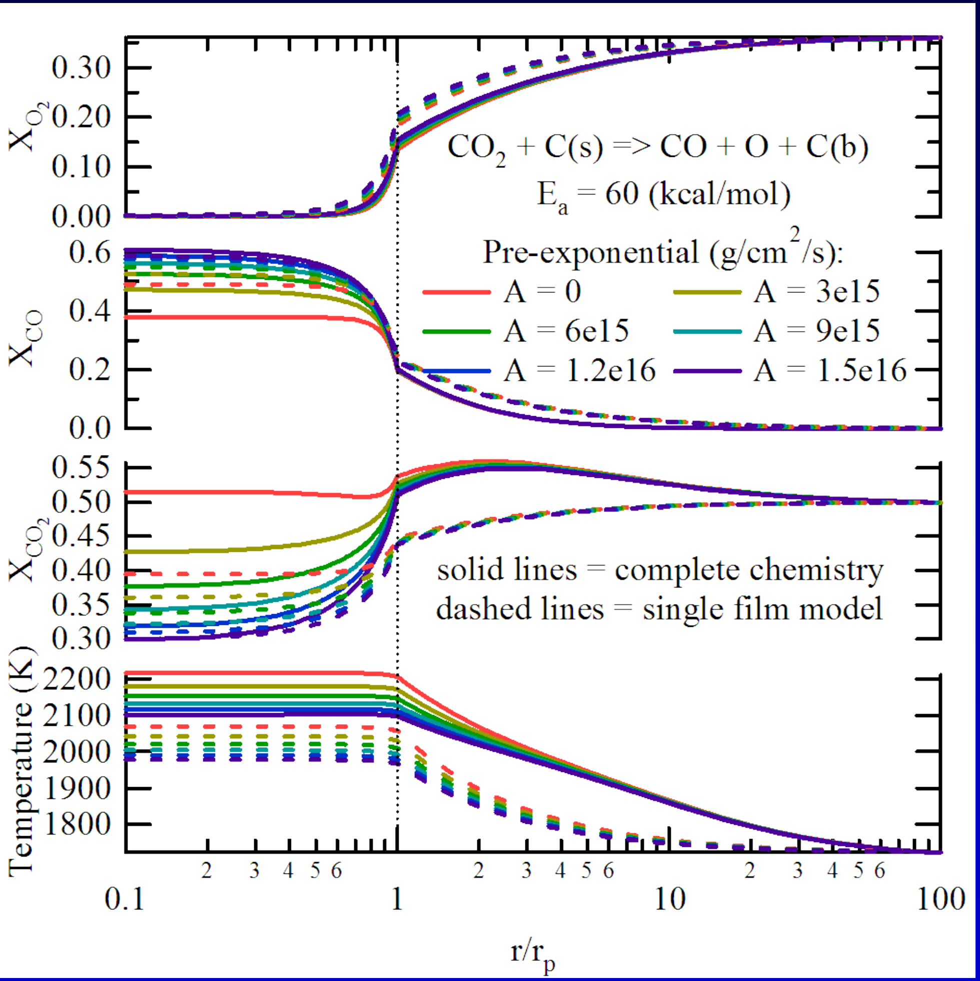
Influence of CO_2 Gasification Reaction during Oxy-Fuel Combustion

- **Widely varying conjecture over possible contribution of CO_2 gasification of coal char during oxy-combustion**
  - many: reaction rate is too small to be important
  - some: CO_2 gasification leads to improved burnout
  - effect of this *endothermic* reaction on overall conversion rates is unclear when coupled with oxidation
- **Actual kinetic rate of this reaction under combustion temperatures (1600-2000 K) is somewhat uncertain**
- **Approach:**
  - SKIPPY simulations, based on literature review of best avail. rates
  - experimental measurements (burnout and particle T) in LEFR in nearly pure CO_2

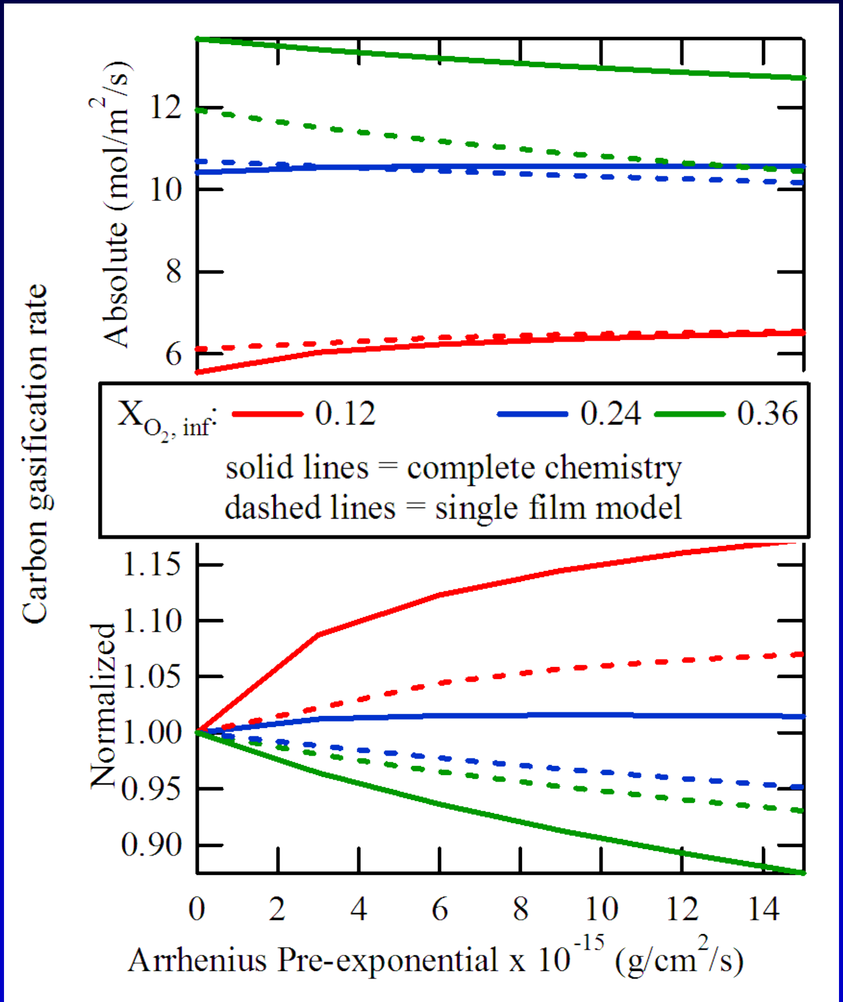
SKIPPY Simulation Results for CO_2 Gasification Reaction, 12 vol-% O_2



SKIPPY Simulation Results for CO_2 Gasification Reaction, 36 vol-% O_2



SKIPPY Simulation Results for CO_2 Gasification Reaction, Overall Burnout Rate



Conclusions

- In moderate temperature environment (1150 K), oxygen concentration and presence of CO_2 both influence ignition of dense stream of pc particles
- Oxygen effects on ignition are stronger than CO_2 effects
- SKIPPY simulations show boundary layer conversion of CO is significant for char particles larger than 60 µm, leading to uncertainty in applicability of single-film analysis
- SKIPPY simulations show CO_2 gasification likely fast enough to decrease char combustion temperature during oxy-fuel combustion, increasing char conversion in low O_2 environments and decreasing char conversion in high O_2 environments

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End of Presentation

Questions?

Conceptual Model of Single-Particle Ignition and Devolatilization