SAND2010-8205P

Effect of Particle Size on Oxy-Fuel Combustion Rates of Pulverized Coal

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Project Update
Salt Lake City, UT, USA
November 11, 2010

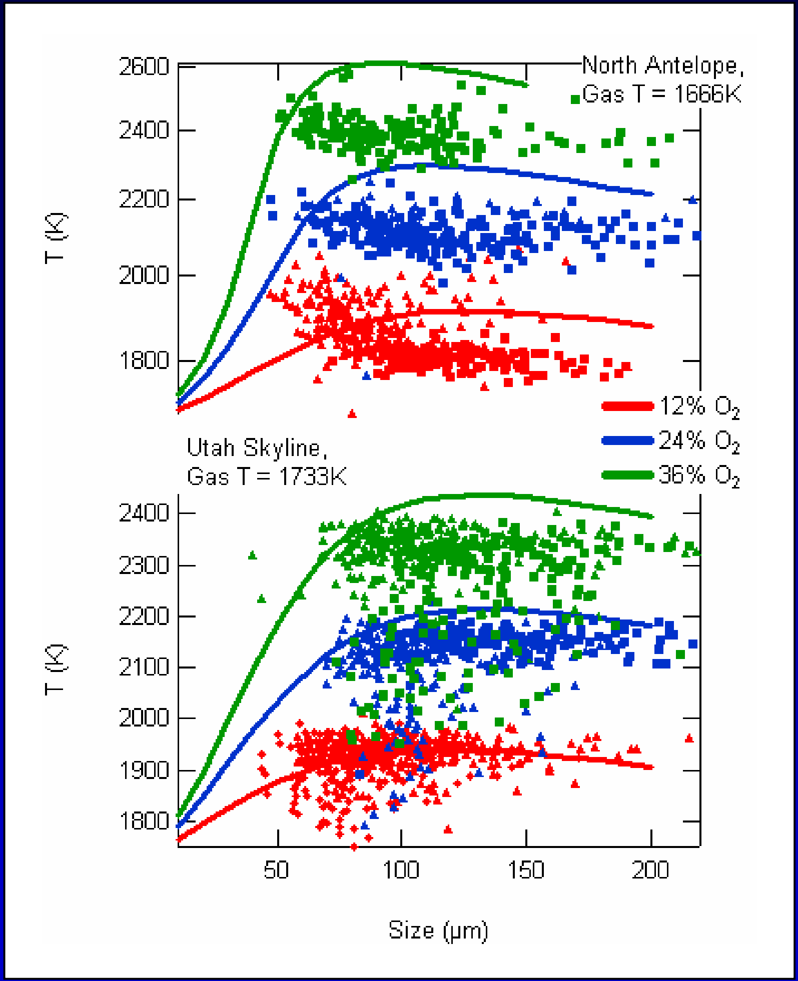
Approach and Research Goals

- Perform experiments in Sandia's optical entrained flow reactor for project coals, measuring char combustion temperature/particle size statistics in oxyfuel environments
- Apply SKIPPY (= Surface Kinetics in Porous Particles) to understand contribution of boundary layer reactions and CO_2 and H_2O gasification reactions to measured char particle temperatures for different O_2 concentrations and particle sizes
- Derive appropriately detailed, CFD-friendly char combustion model that accounts for vagaries of oxyfuel combustion (char gasification reactions; boundary layer conversion of CO)

Status as of Last Review (Jan. 2010)

- Char combustion temperature/particle size statistics collected in oxyfuel environments with 12 vol-% < [O_2] < 36 vol-%
- Single-film modeling of data with *n*th-order Arrhenius oxidation model shows overprediction of particle T with increasing O_2 levels
- SKIPPY simulations with base oxidation model shows even larger overprediction with increasing O_2 levels, unless substantially decrease assumed surface area density
- Conclusions:
  - CO_2 gasification reaction may need to be included (in both modeling approaches)
  - measurements should be made on prepared char particles to delineate effect of devolatilization environment on effective char surface area (i.e. reactivity)

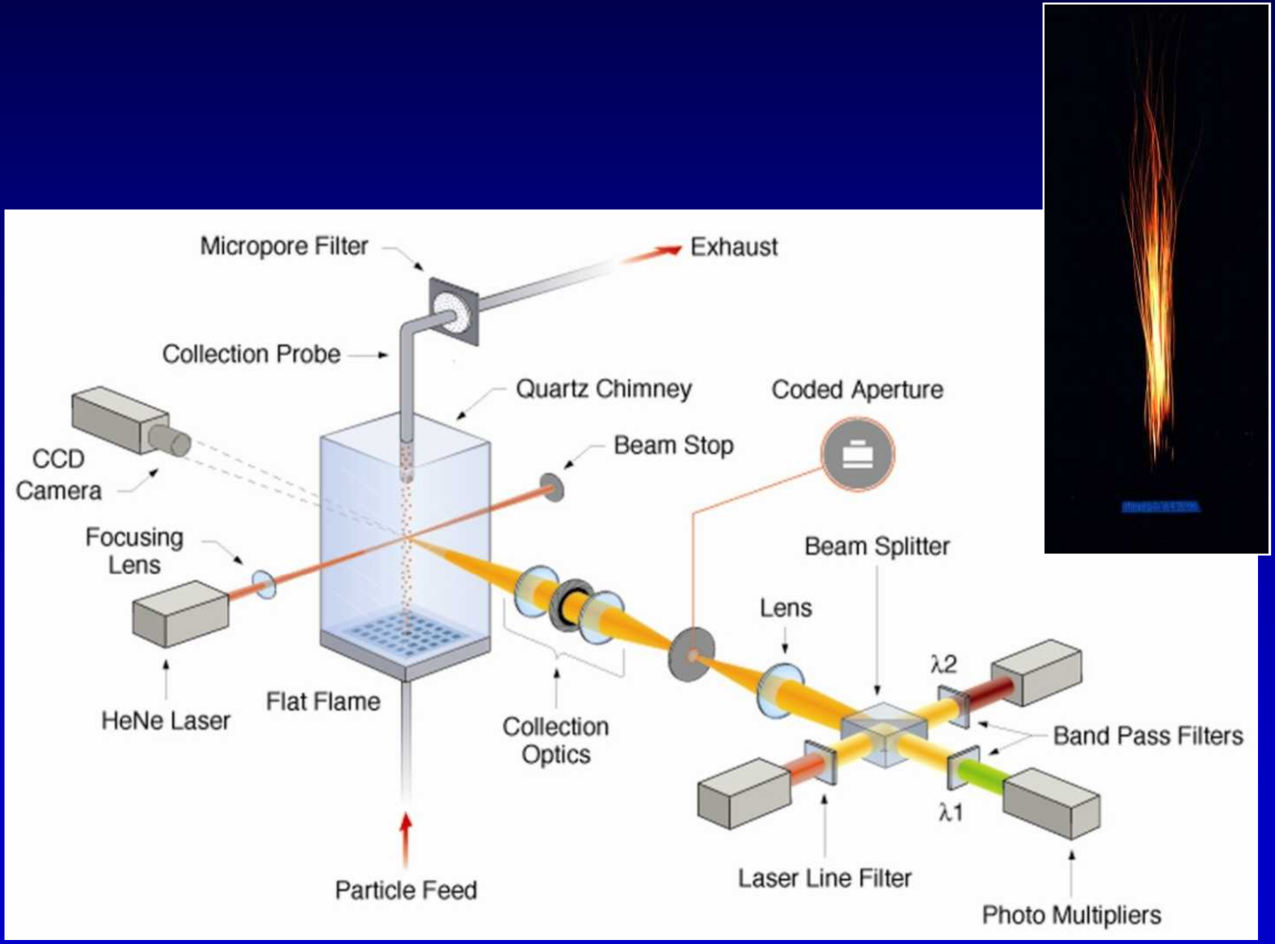
Previous Single-Film Modeling Results



New Work

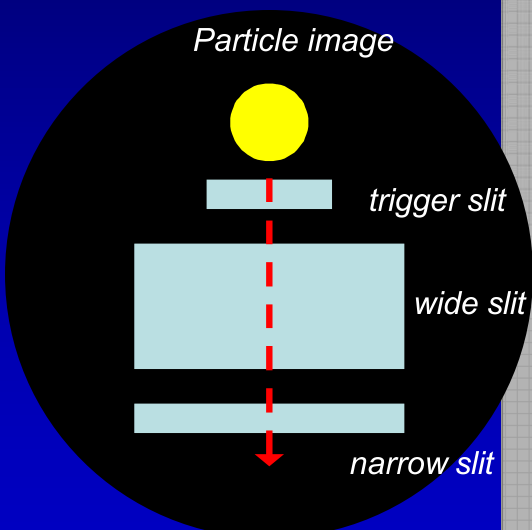
- Char combustion temperature/particle size statistics collected in N_2 environments with 12 vol-% < [O_2] < 36 vol-%, so models have baseline dataset with minimal gasification reactions to calibrate oxidation model
- Char particle combustion data collected for partial set of conditions using char particles prepared in drop tube reactor
- CO_2 gasification reaction, and then steam gasification reaction, added to SKIPPY model simulations (16 vol-% H_2O in experimental furnace environment)
- Single-film, *n*th-order Arrhenius char combustion model fit to experimental data for different values of *n*

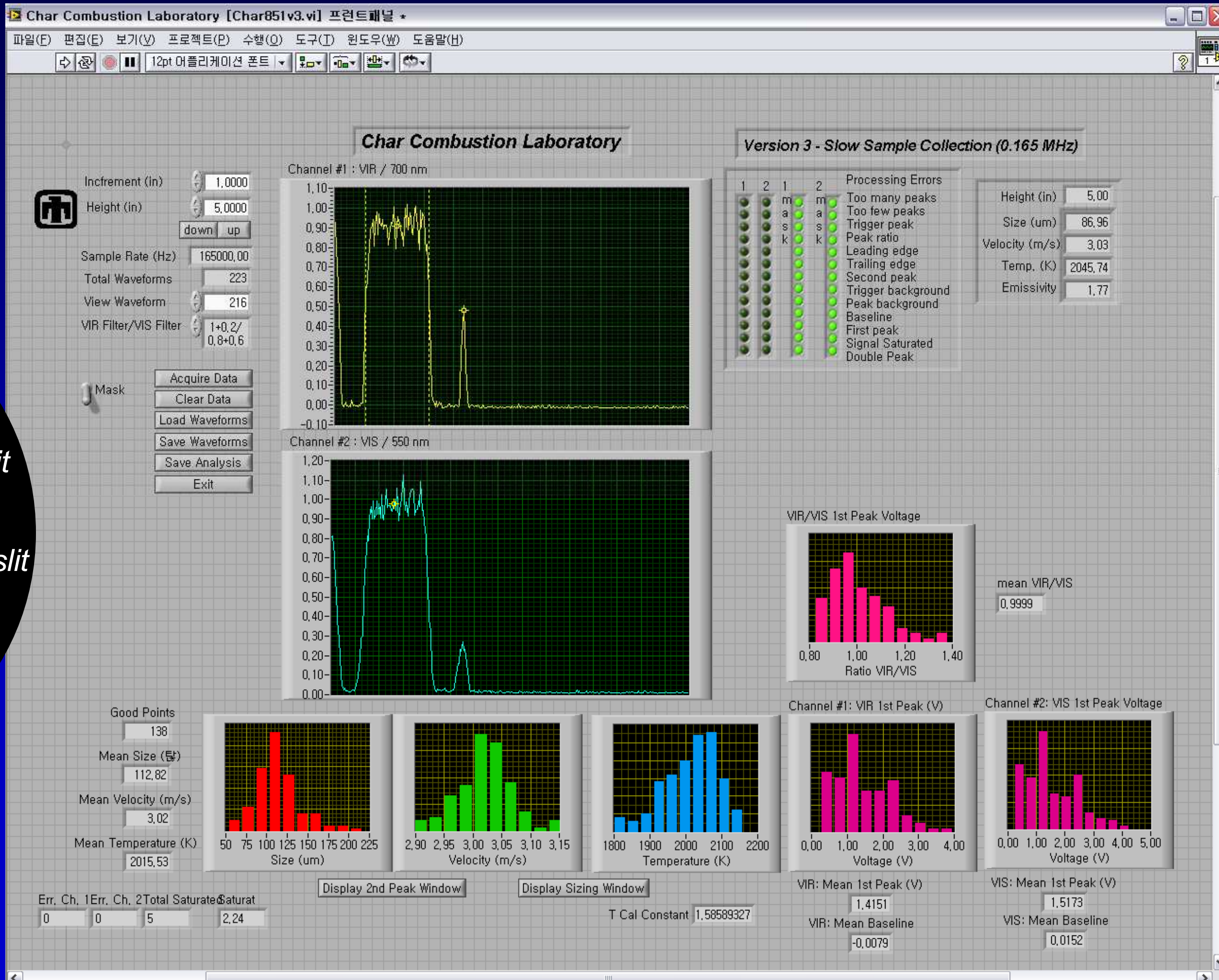
Experiments use a combustion-driven optical entrained flow reactor



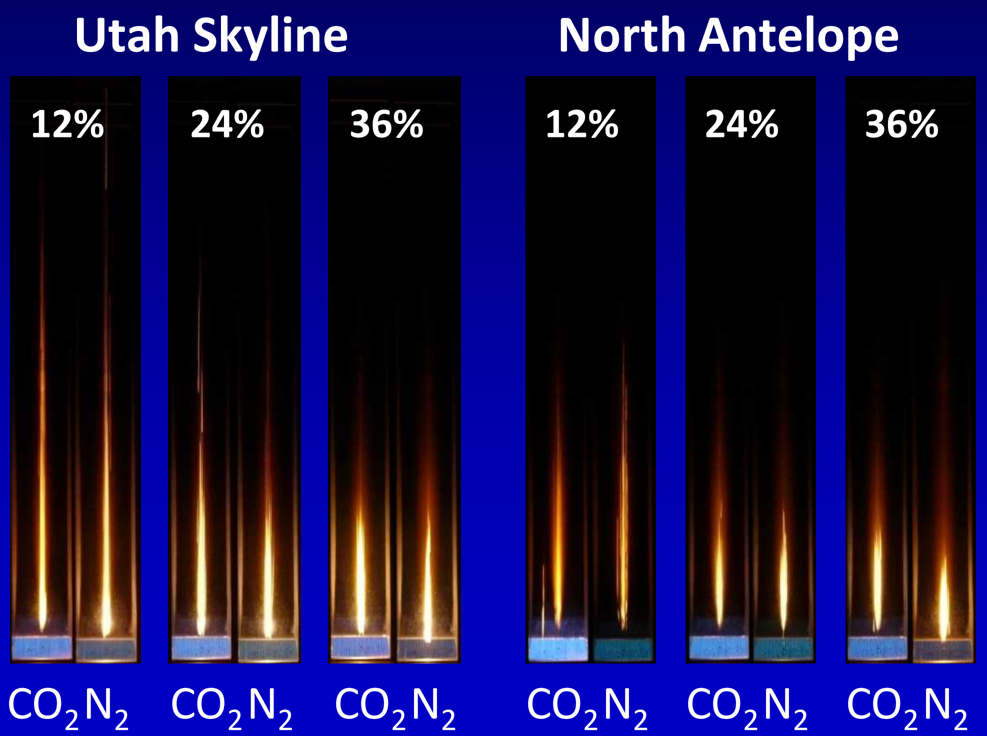
- 1 atm
- compact, diffusion-flamelet burner
- coal or char particles introduced along centerline
- quartz chimney
- coded-aperture, 2-color pyrometry diagnostic for char size, T, and velocity
- laser-triggered ICCD for single particle imaging

Particle-sizing pyrometry in the laminar entrained flow reactor



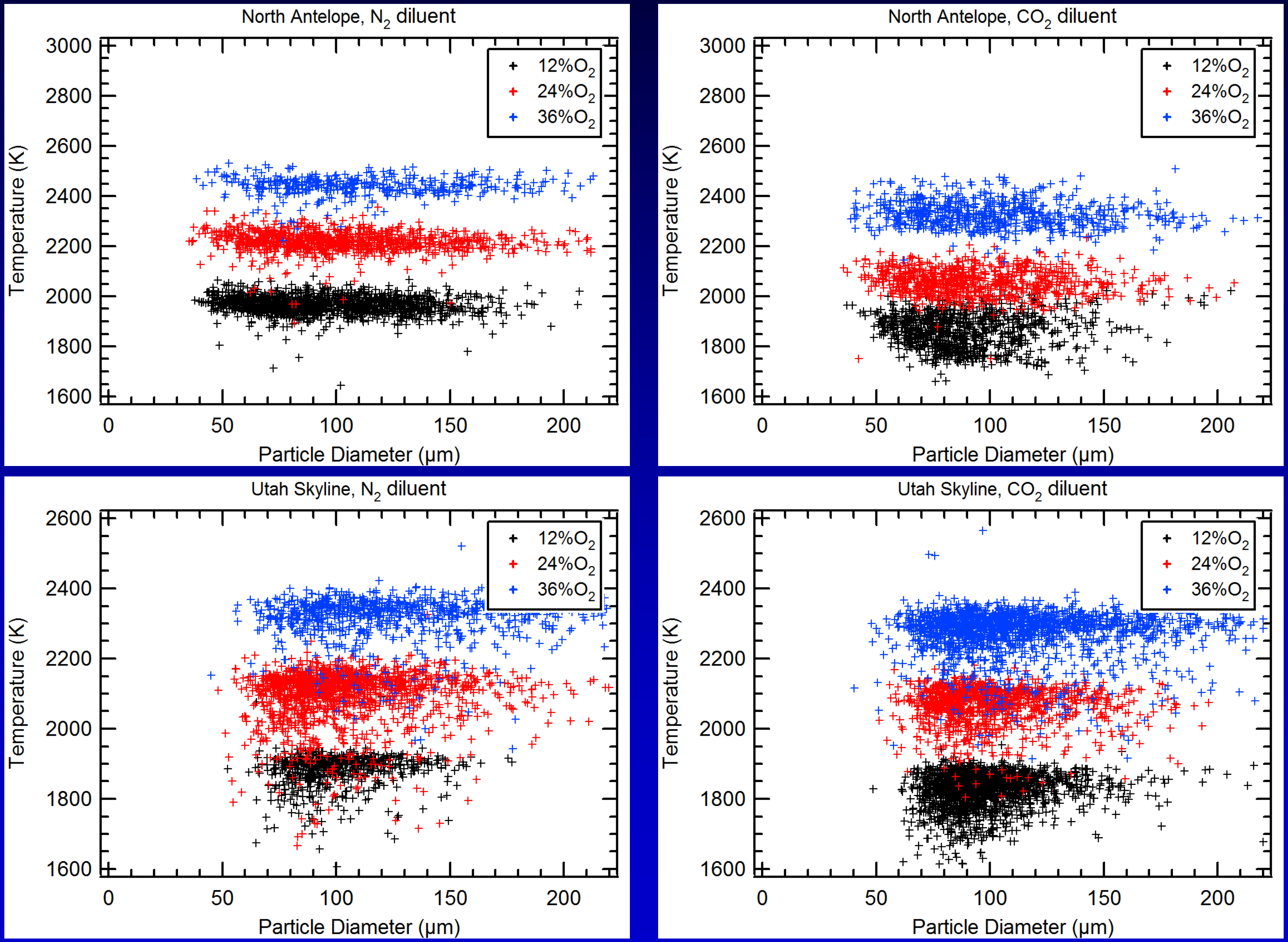


A constant-speed coal feeder ensured steady introduction of pre-sieved coal

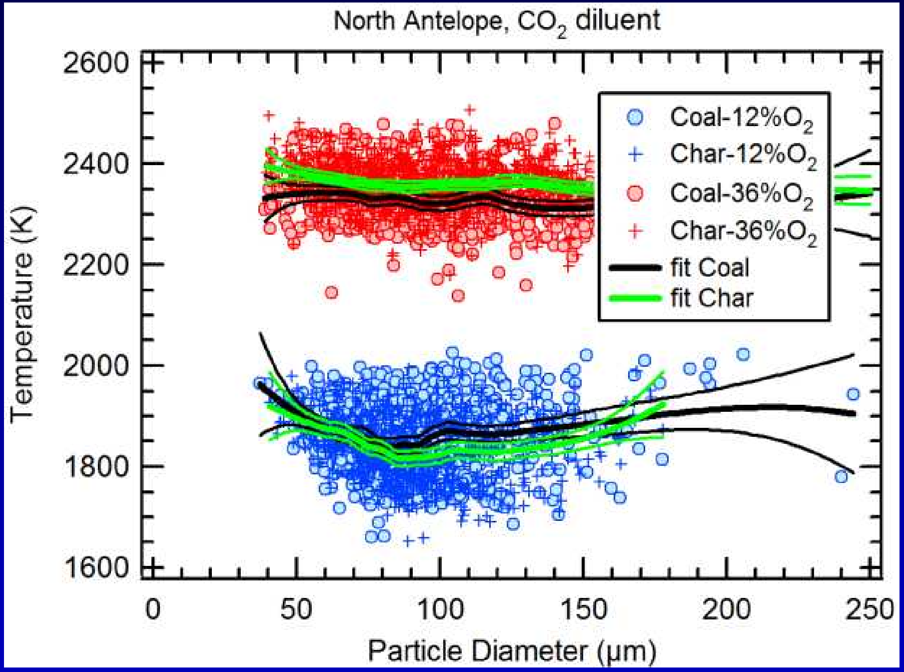


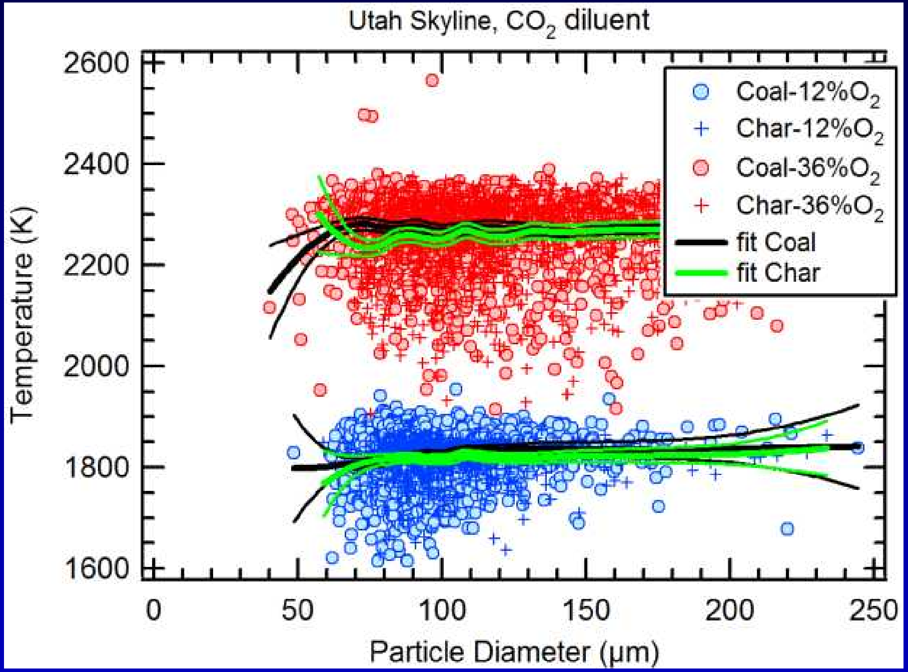
- **2 Coal ranks**
  Bituminous (Utah Skyline)
  Subbituminous (North Antelope)
- **75 – 106 µm size fraction**
- **3 Oxygen concentrations**
  12, 24, and 36 vol-%
- **~ 1700 K** furnace temperature
- **16%** moisture
- **60 slpm** overall product flow rate
- **< 1g/hr** coal feed rate
- N_2 or CO_2 diluent gas

Particle-sizing pyrometry data (coal feed)



Particle-sizing pyrometry data comparison for raw coal and prepared char feed



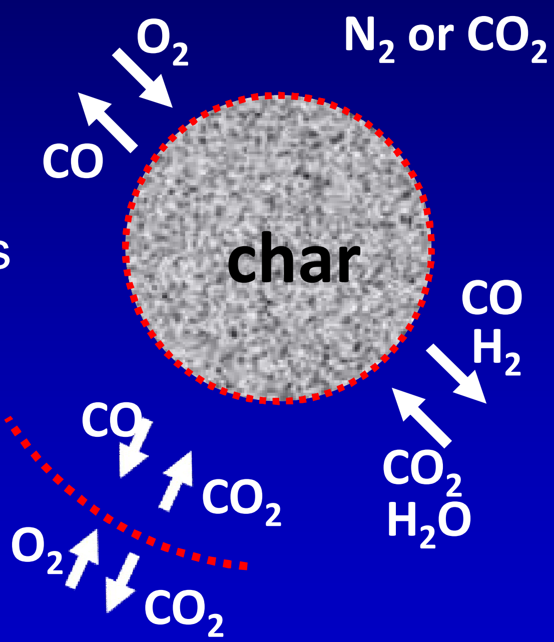


- Chars prepared by feeding 75–150 µm size-classified coal into drop tube reactor operating at 1200 deg C with 3% O_2 in N_2; 0.25 s res. time
- Chars collected in water-cooled, gas-quench sampling probe, separated in cyclone, then sieved to 75–106 µm for combustion experiments

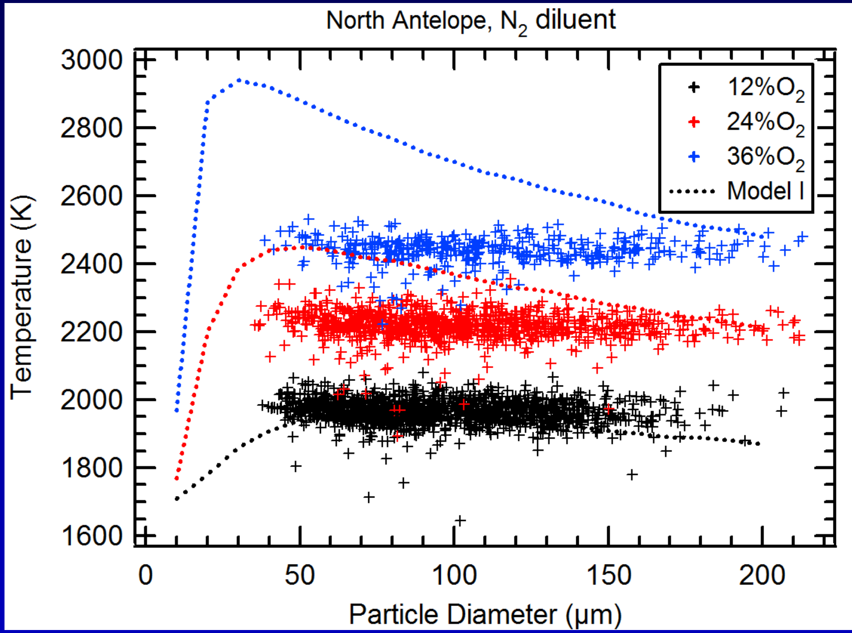
SKIPPY calculates temperature and species profiles for steady-state burning of porous char particles

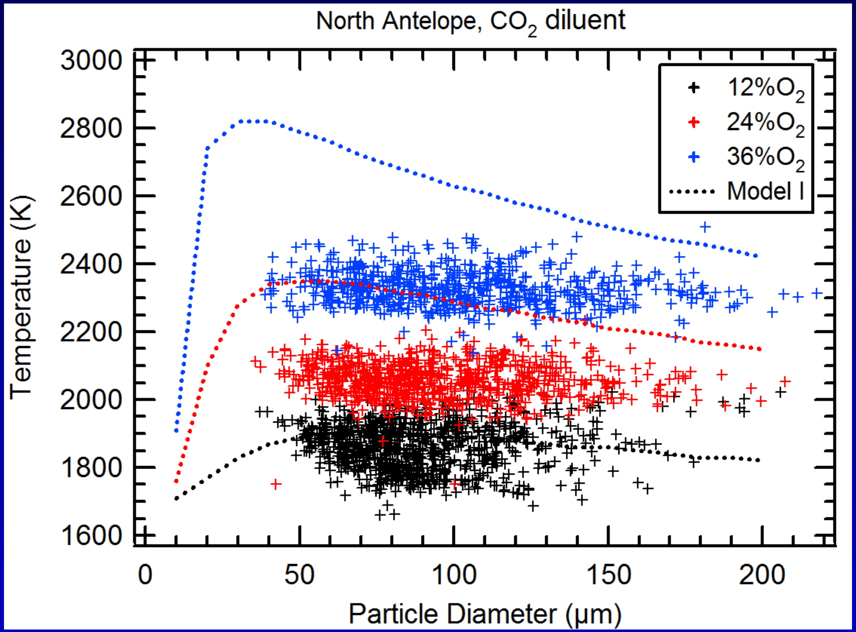
- **SKIPPY = Surface Kinetics in Porous Particles**
- **Utilizes CHEMKIN II surface kinetic approach**
- **Our simulations:**
  - GRI-MECH 3.0 gas-phase kinetics
  - typical char properties, experimental conditions

Reaction	*A* (g/cm² s)	*E* (kJ/mol)
Heterogeneous oxidation:		
(R1) C_s + O_2 => CO + O_s	3.3E+15	167.4
(R2) O_s + 2C(b) => CO + C_s	1.0E+08	0.
(R3) C_s + O_2 => O_2_s+ C(b)	9.5E+13	142.3
(R4) O_2_s + 2C(b) => C_s + CO_2	1.0E+08	0.
CO_2 gasification reaction:		
(R5) C_s + CO_2 => CO + O_s + C(b)	3.60E+15	251.0
Steam gasification reaction:		
(R6) C_s + H_2O => H_2 + O_s + C(b)	4.35E+14	222.8



SKIPPY predicted and measured single particle temperatures

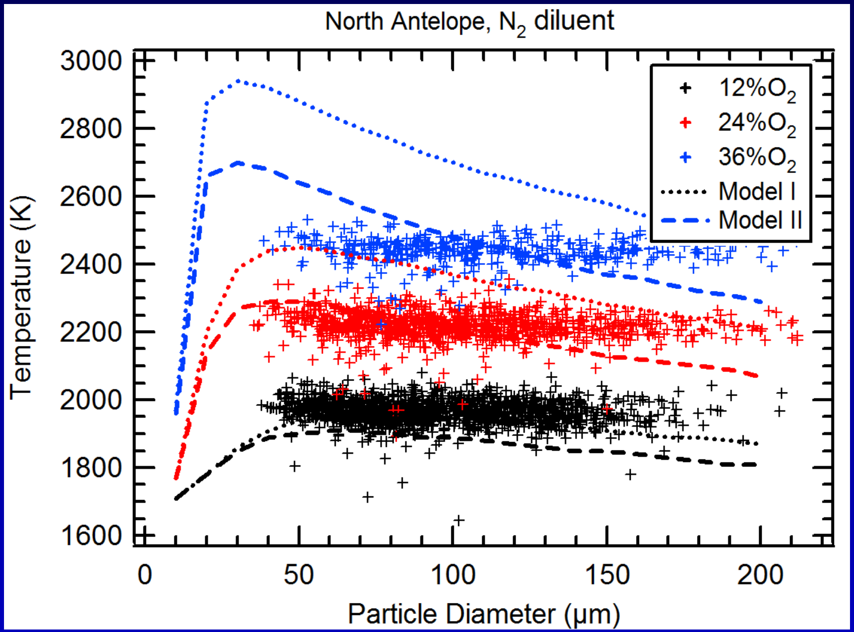


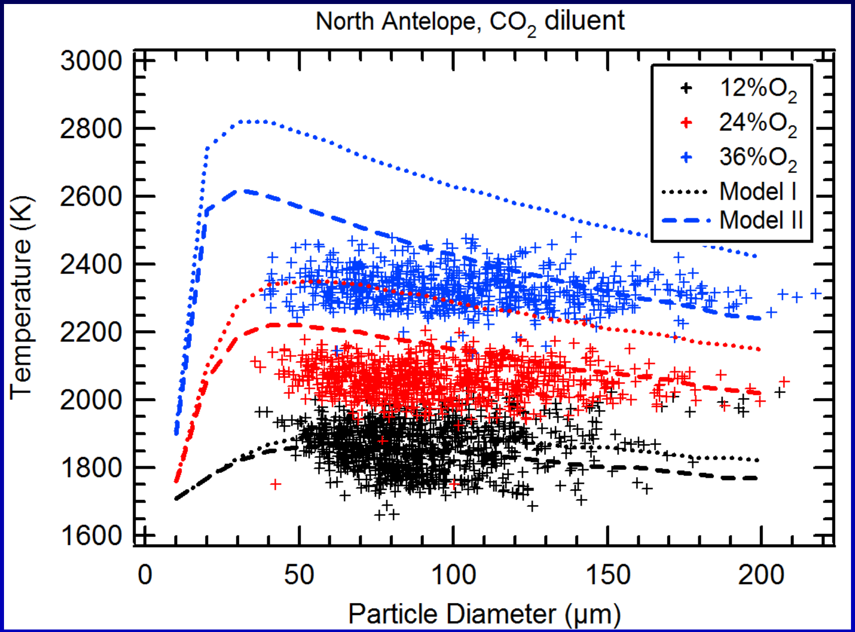


Use constant assumed surface area density: 10 m^2/g

- Model I: char oxidation only

SKIPPY predicted and measured single particle temperatures

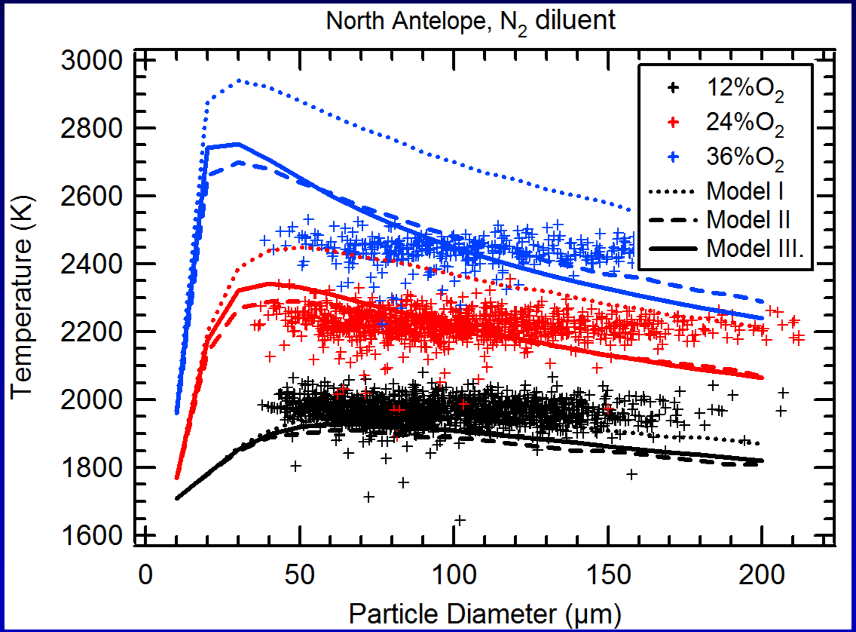


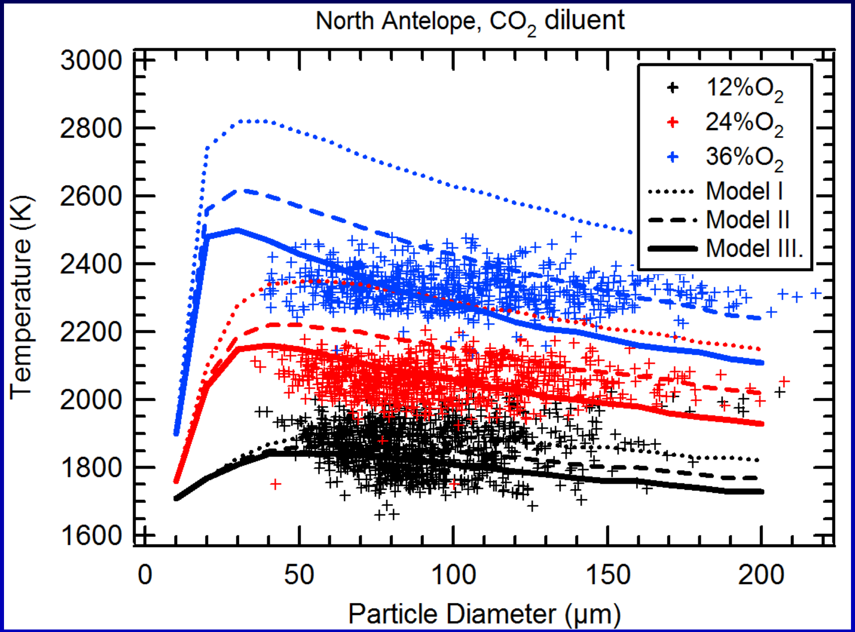


Use constant assumed surface area density: 10 m^2/g

- Model I: char oxidation only
- Model II: char oxidation + steam gasification

SKIPPY predicted and measured single particle temperatures





Use constant assumed surface area density: 10 m^2/g

- Model I: char oxidation only
- Model II: char oxidation + steam gasification
- Model III: char oxidation + steam gasification + CO_2 gasification

Single-film model prediction of particle temperature as function of particle size

- **Use single-film *n*th-order Arrhenius kinetic rate expression for oxidation**

$$k_s = A\exp\left(-E/RT_p\right) \qquad q = k_s p_{O_2,s}^n$$

- **Solve instantaneous energy balance for a reacting particle:**

$$\frac{d_p \rho_p C_{v_p}}{6}\frac{dT_p}{dz} = -\varepsilon\sigma\left(T_p^4 - T_w^4\right) - \frac{2\lambda}{d_p}\left[\frac{\kappa/2}{e^{\kappa/2}-1}\right]\left(T_p - T_g\right) + q\Delta h$$

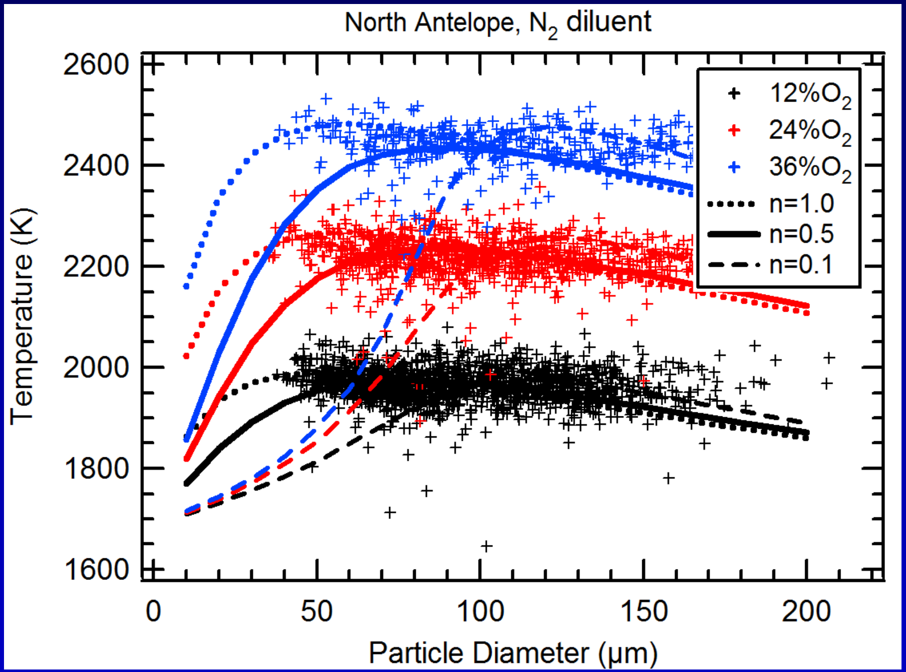
  - Determine reaction heat release using Tognotti CO_2/CO production ratio

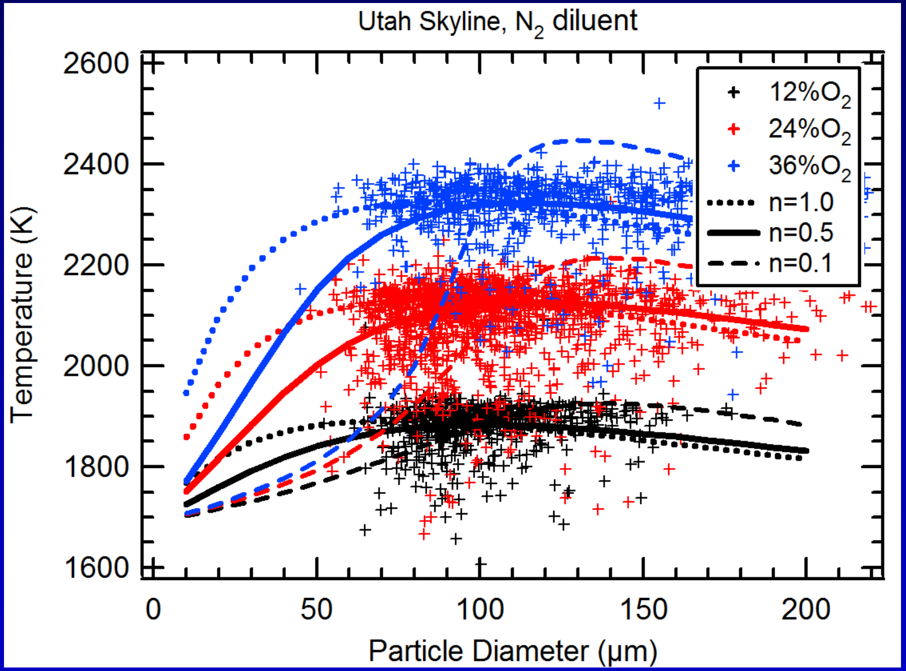
$$CO_2/CO = 0.02\,p_{O_2,s}^{0.21}\exp\left(3070/T_p\right)$$

  - $dT_p/dz = 0$

Single-film oxidation simulation results

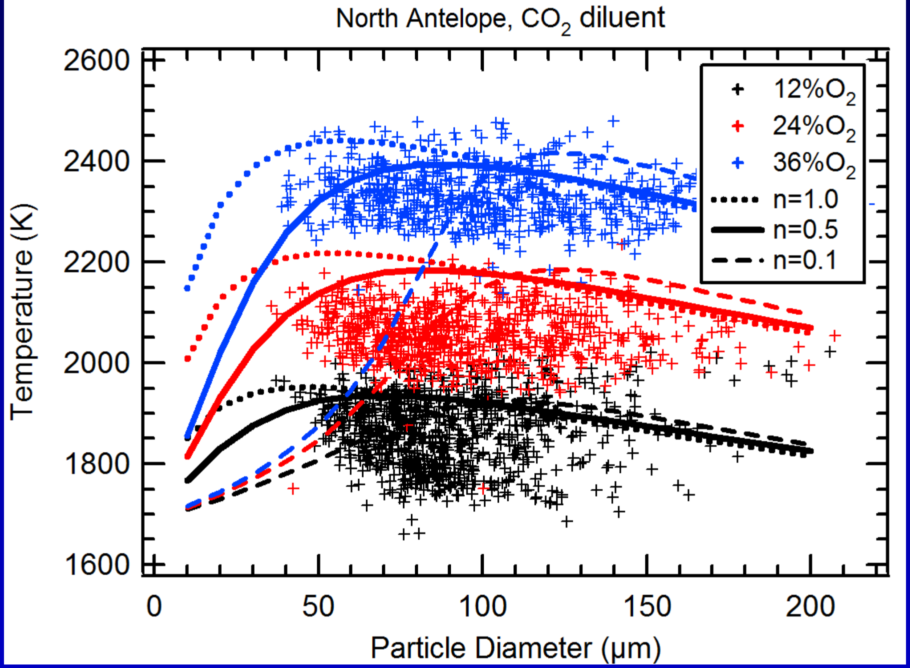
N_2 diluent

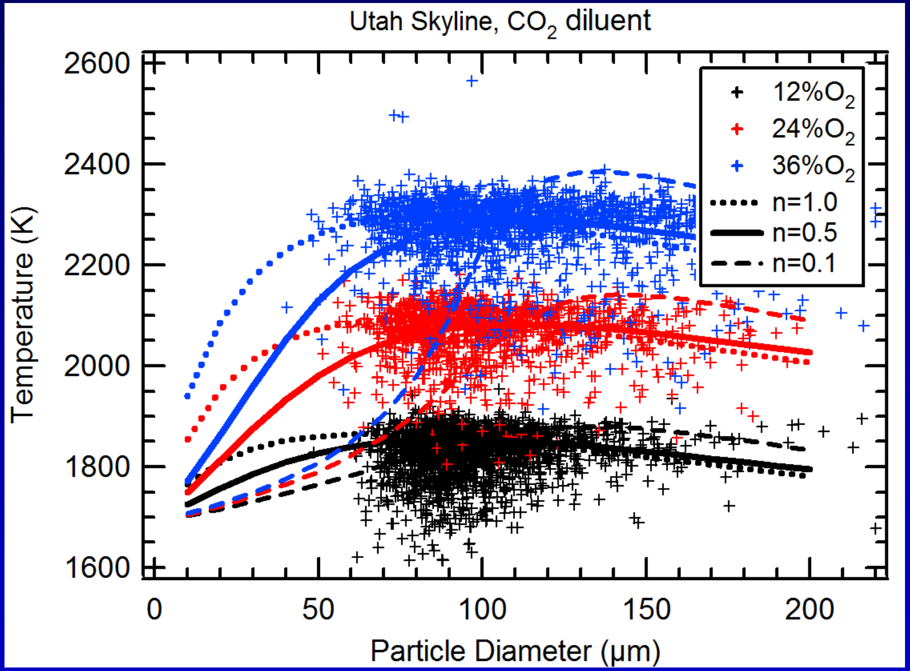




Single-film oxidation simulation results

CO_2 diluent





Single-film oxidation simulation results

*n*th-order fit parameters

	North Antelope:	
Fixed n =	A (kmol/m²·s)	E (kJ/mol)
1	0.44	0.0
0.5	0.67	36.5
0.1	1.70	76.3
	Utah Skyline:	
1	0.39	12.0
0.5	1.05	51.8
0.1	2.31	83.5

Non-physical fit parameters (e.g. E_a << 80 kJ/mol) suggest important physics missing from model; danger in extrapolating beyond fit conditions

Conclusions

- North Antelope coal shows small sensitivity of reactivity to devolatilization of isolated particles in flow reactor with different O_2 levels (whereas Utah Skyline is insensitive) – char combustion studies with preformed char particles recommended (in general)
- SKIPPY simulations show both steam and CO_2 gasification reactions need to be included to get acceptable agreement in measured and calculated burning rate (i.e. char particle temperature) for both conventional and oxyfuel combustion conditions
- Single-film simulations with *n*th-order Arrhenius oxidation show decent agreement with data as long as n = 0.5 or greater. Fits for N_2 diluent data give high prediction for North Antelope data during oxyfuel combustion. Activation energies for these fits with large n are very low.

Next Steps

- Investigate effect of assumed surface area density in SKIPPY on variation of predicted char particle temperatures as function of particle size
- Implement single-film model that includes char oxidation and gasification reactions – compare optimized predictions against data
- Develop recommendations for best CFD-friendly way to model char combustion rates of North Antelope and Utah Skyline coals during oxyfuel combustion

Acknowledgments

Brian Haynes, Univ. of Sydney, for graciously providing his SKIPPY code

Research was sponsored by U.S. DOE under Award Number DE-NT0005288, with Reaction Engineering International (REI) as the project lead. This project is managed by Mr. Timothy Fout of the National Energy Technology Laboratory (NETL).

Thank you!