SAND2011-2700P

Research on Coal Combustion and Gasification Science at Sandia National Labs

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North Antelope mine with unit trains, Powder River Basin, WY

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Project Timeline and Funding

- Started FY02
- Funded at $400K/yr (flat funding profile)
- Funds spent to-date $3.6M

Research Thrusts (Tasks)

- Development of Advanced Combustion/Gasification Reactor (FY02)
- PC Char-NOx Formation and NOx Reburn (FY02-FY04)
- Advanced Combustion of PC (FY02)
- Char Kinetics for Oxy-Fuel Combustion (FY03-FY08)
- NOx Formation and Reduction during Oxy-Fuel Combustion (FY05-FY08)
- Pressurized Char Gasification Kinetics (FY08-FY12)
- Pressurized Char Combustion Kinetics (FY09-FY13)

Motivation: Development of Advanced Comb./Gasif. Reactor

- Existing SNL coal research facilities limited to 1 atm operation – inappropriate for investigating gasification or pressurized combustion phenomena
- In conjunction with funds provided by DOE-EE Office of Biomass Programs, opportunity arose for design and construction of lab-scale pressurized flow reactor for combustion and gasification studies

Approach: Development of Advanced Comb./Gasif. Reactor

- Specify desired experimental capabilities
- Collect design and operational information on all similar existing or recently decommissioned lab-scale rigs with controlled temperatures
- Design, construct, and conduct experiments on heavily instrumented pressurized test rig to evaluate effective thermal conductivity of alumina/zirconia insulation at high temperatures
- Complete final design, procure parts, and construct reactor, in consultation with SNL ES&H experts

Results: Development of Advanced Comb./Gasif. Reactor

- Determined desired capabilities:

  1 – 20 bar pressure

  300 – 1600 K

  0.2 – 5.0 s residence time

  multiple fixed gases and steam flow

  turbulent pipe flow

  traversable particle/gas collection

  optical access

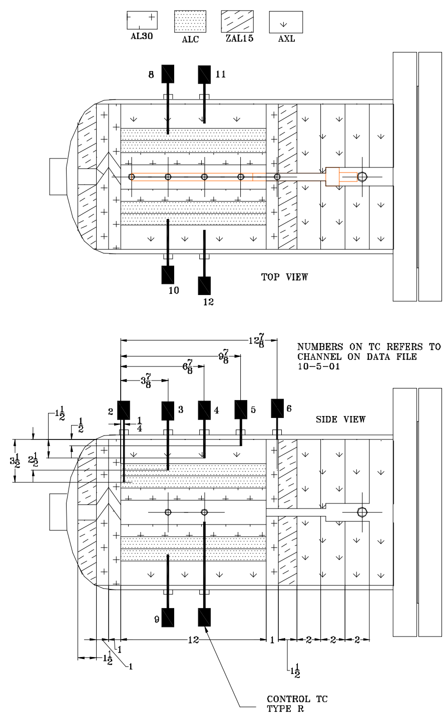
- Evaluated related rig designs:

Reactor Owner/Location	Reactor Name	Important Features	Literature Sources
IPST Atlanta, GA	PEFR	entrained flow reactor: 1-80 atm, 873-1773 K, 3" x 66" reactor tube, traversable water-cooled sampling probe, 4 optical sampling heights	Hansen et al., '95
VTT Jyväskylä, Finland	PEFR	entrained flow reactor: 1–20 atm, 1000–1700 K, 2.4" x 66" reactor tube, hot-water traversable sampling probe, 3 optical sampling heights	Aho et al., '95
IVD Stuttgart, Germany	PEFR	entrained flow reactor: 1-16 atm, up to 1873 K, 2.8" x 69" reactor tube, hot-water traversable sampling probe, 3 optical sampling heights	Reichelt et al., '98 Reichelt et al., '00
CRC Australia	PDTF	drop tube furnace: 1-16 atm, 873-1673 K, 3" x 50" reactor tube, traversable injection, water-cooled sampling probe, 4 optical sampling heights	Ouyang et al., '98 Yeasmin et al., '99
BYU Provo, UT	HPCP	drop-tube reactor: 1–15 atm, 1000–1700 K, 2" x 20" reactor tube, 1 optical location	Monson, '92 Monson and Germane, '93 Bateman et al., '95
PSU University Park, PA	HEF	entrained flow furnace: 6-70 atm, up to 1373 K, 1.6" x 20" reactor tube, water-cooled sampling probe	Lee et al., '91
NETL Morgantown WV	CDER	entrained reactor: 1-12 atm, up to 1720 K; 2" x 25" reactor tube, 2 optical sampling heights	Anderson et al., '90

inactive

Results: Development of Advanced Comb./Gasif. Reactor

- Literature review revealed that existing research reactors suffer from unexpectedly high heat loss to pressure shell when pressurized
- An insulation test vessel was designed and operated to identify the predominant mechanism of unanticipated heat loss



- Heat transfer analysis demonstrated slightly higher thermal conductivity of insulation at pressure; also demonstrated strong buoyancy-driven heat transfer at pressure

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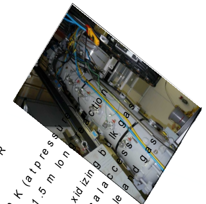
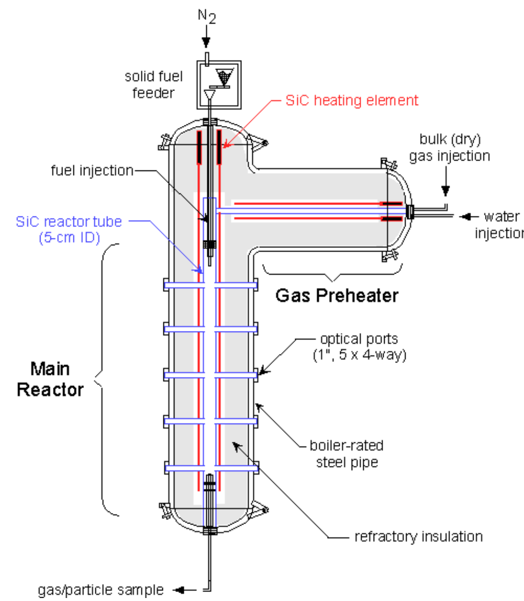
Major Accomplishme... Dev..op..nen... Advanced Comb./... ...tor...

- New experimental rig de... ...d inst... care taken to minimize ... buoyan... ...ithin the pressure vessel
- Design parameters:

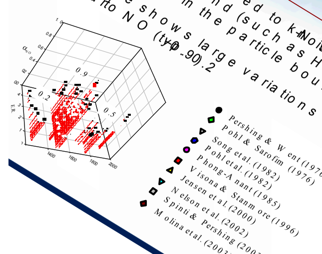


- ✓ turbulent EFR
- ✓ 1–20 bar
- ✓ 300–1600 K (at pressure)
- ✓ 5 cm ID x 1.5 m long tube
- ✓ reducing or oxidizing bulk gas
- ✓ 5-point 4-way optical access
- ✓ traversable particle and gas sampling system



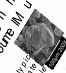
Motivation - NO Formation and Char-N Reburn

- CFD modelers need to know whether char-N is released as NO, or compounds (such as HCN) and the fate of these compounds in the particle boundary layer
- Literature shows large variations in reported fractional conversion of char-N to NO (typ. 0.2-0.9)



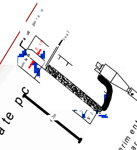
Approach-NO Formation on Char and Reburn

- Develop technique for generating representative high heating rate pc char particles from Multi-Fuel Combustor (MFC)

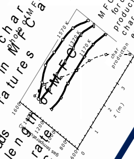


SEM of typical MFC generated char particle

- Establish flat T profiles for char oxidation experiments in MFC at 3 characteristic temperatures
- Implement in-line gas sampling along length of MFC
- Determine NO reaction rate with pc char in MFC



experimental setup for pc char generation and collection



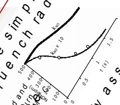
MFC T profiles for char production and char oxidation experiments

Approach – NOx Formation and Char Reburn

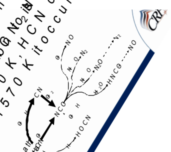
- Develop computational codes for s[illegible] particle boundary layer chemistry in MFC with burning [illegible] gas [illegible]es, allowing detailed model compa[illegible] experiment
- Develop novel technique for s[illegible] gas phase radical species in MFC, [illegible] discrimination of alternative NOx formation [illegible] to
- Measure NOx production rate from char as function of gas temperature, background NO, and with radical scavenging on/off
- Compare experimental results with models, for different presumed char-N release chemistry (NO or HCN)

Major Accomplishments: NO Formation and NOx Reburn

- Determined that gaseous HCN addition is the simplest, safest, and most effective approach to quench radical reactions in MFC
- Determined that NO–char reaction rate proceeds at 10x average literature rate,
- Determined that char-N is predominantly released as HCN, not as NO, as commonly assumed
- Determined that HCN oxidation to NO or N_2 is dependent on T. At 1070 K, HCN oxidation occurs in bulk gas, whereas at 1570 K it occurs in particle boundary layer



Measured and modeled NO during char... at 1490 K



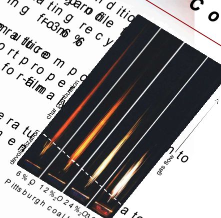
Reaction path analysis of HCN... NO formation at 1170 K

Motivation: Advanced Combustion of P

- Oxygen-enriched combustion lowers gas cleanup costs and can improve furnace efficiency
- With flue gas re-cycle, oxy-combustion allows for CO_2 capture from a high efficiency furnace
- The characteristic combustion properties of pc char in elevated O_2 and elevated CO_2 environments need to be determined for accurate CFD modeling

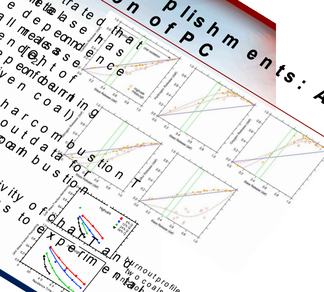
Approach: Advanced Combustion of PC

- Establish EFR operating conditions ... identical temperature profile ... N_2 and O_2/CO_2 environments simulating recy[cle] conditions with varying O_2 from 6% to 36%
- Incorporate char temperature ... gas mixture transport properties ... computational code for film ... char reactivity
- Measure char particle temperature ... burnout profiles, and element[al] ... for 6-36% O_2 in N_2 and CO_2



Major Accomplishments: Advanced Combustion of PC

- Demonstrated that element release has same dependence on overall mass release (i.e. independent of rate of burning for a given coal)
- Provided char combustion and coal burnout data for comparison with combustion models
- Analyzed sensitivity of char burnout predictions to experimental uncertainties



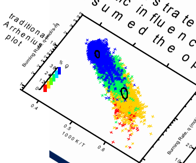
T and burnout profiles for two coals as a function of bulk O

Motivation: Kinetics for Oxy-Fuel Combustion

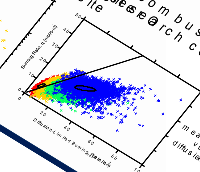
- Accurate CFD models of oxy-fuel combustion burners and boilers requires quantitative coal ignition delay and char combustion kinetic rates in these unique environments
- With localized oxygen addition in many designs, coal ignition and char combustion occur over a wide range of O_2 levels
- Flue gas recirculation, often presumed to be wet, means ignition and char combustion occur in environments with elevated CO_2 and possibly H_2O levels, introducing changes in heat capacity, gas transport properties, and possibly the influence of char gasification reactions

Results: Kinetics of Oxy-fuel Char Combustion

- Developed non-linear, discrete methodology for fitting single film char combustion kinetics models (nth order, Arrhenius, Langmuir-Hinshelwood, etc.) to experimental data from optical entrained flow reactor
- Demonstrated that char combustion occurs under increasing kinetic influence for higher O_2; research community had assumed the opposite



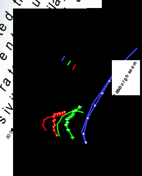
traditional Arrhenius plot

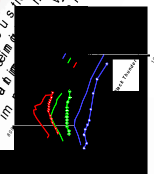


measured rate vs. diffusion limit

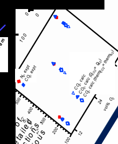
Results: Kinetics of Oxy-Fuel Combustion

- Demonstrated that char combustion temperature is lower in CO_2 environment ... char oxidation kinetic rate ... lower O_2 diffusivity ...





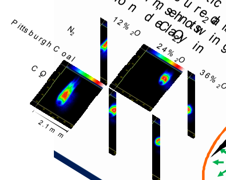
temperature rise of char particles in flow reactor

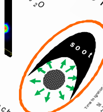


comparison of data with detailed model predictions including fictitious CO_2 properties

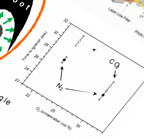
Results: Kinetics of Oxy-Fuel Combustion

- Single particle imaging performed using intensified CCD camera with high magnification optics
- Characteristic coal particle ignition delays measured in N_2 and CO_2 environments, showing longer ignition delay in CO_2



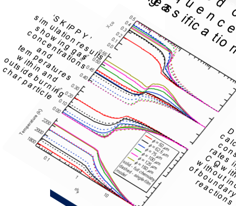


schematic of single particle volatiles fed diffusion flame

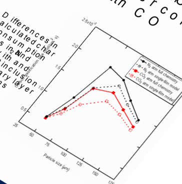


Major Accomplishments: Kinetics for Ox Combustion

- Traditional char oxidation kinetic models shown to fail for elevated oxygen levels in CO_2 environments as a consequence of boundary layer conversion of CO and because they ignore gasification reactions with CO_2



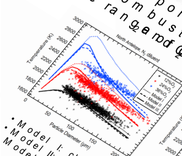
'SKIPPY' simulation results showing gas concentrations and temperatures within and outside burning char particle

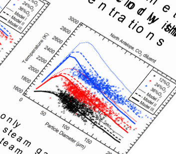


Differences in calculated char consumption rates in N_2 and CO_2 with and without inclusion of boundary layer reactions

Major Accomplishments: Char Kinetics for Oxy-Combustion

- Demonstrated that including char gasification reactions, with literature-supported rate parameters, allows accurate prediction of char combustion behavior using kinetic model for wide range of O_2 and CO_2 concentrations



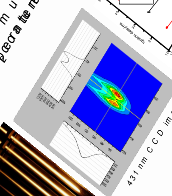


- Model I: char oxidation only
- Model II: char oxidation + steam gasification
- Model III: char oxidation + steam gasification + CO_2 gasification

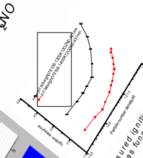
Major Accomplishments: FY 10 Kinetics for Oxy Combustion

- Determined ignition delay of coal streams with a wide range of particle densities for multiple coals, in several size cuts, over range of gas temperatures, and in O_2/CO_2 and O_2/N_2 environments

photos of coal stream ignition and combustion as function of particle loading



431 nm CCD image of ignition



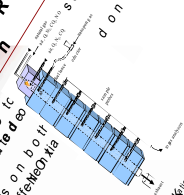
measured ignition delay for different gas T as function of particle loading

Motivation: NOx Formation and Reduction during Oxy-fuel Combustion

- Most studies find significantly lower NOx formation than normal air-fired during oxy combustion (~ 3x lower)
- Contributing factors are clear:
  - ✓ Negligible thermal NOx (absence of N_2 in oxidizer)
  - ✓ Reburning of recycled NOx on coal flame
  - ✓ Reburning of recycled NOx on coal char
- Quantitative assessment of NOx reduction factors in oxy and knowledge of implications for accurate modeling of oxy NOx formation are lacking

Approach: NOx Formation and Reduction during Oxy-Fuel Combustion

- Convert gas supply to MFC to allow operation in simulated oxy-fuel ... ses to ... d on
- Conduct experiments on both pc char particles, to differentiate volatile-N and char-NOx
- Vary background NO (to determine influence of NO on char-N conversion)
- Vary CO_2 concentration (N_2 or CO_2 diluent)
- Keep gas T profile flat at ~ 1320 K (to simplify interpretation and modeling)



Schematic of MFC after modifications for oxy-firing

Results: NOx Formation and Reduction during Oxy-Fuel Combustion

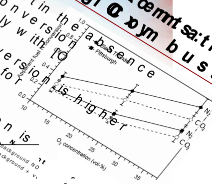
- For comparing NOx data for different coals and different coal injection rates, it is important to convert NOx to a more effective fuel N conversion

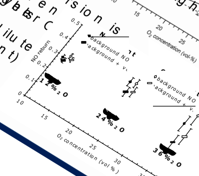
$$\frac{N_{NOx}}{N_{0,fuel}} = \frac{[NOx]\dot{V}\left(\frac{P}{RT}\right)}{\frac{Y_{N,fuel}M_{fuel}}{MW}}$$

- With this conversion, NOx conversion in oxy-fuel is shown to be lower than conversion in air / nitrogen environment

Major Accomplishments: NOx Formation and Reduction during Oxy-Fuel Combustion

- Demonstrated that in the absence of reburning, fuel N conversion increases strongly with [O$_2$]
- Determined N conversion is higher for subbituminous coal than for bituminous coal
- Determined N conversion is reduced in CO$_2$ environment, especially for higher [O$_2$]
- Determined reburn NO (in dilute particle environment) … all conditions





Motivation: Char Gasification Kinetics

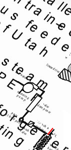
- IGCC technology offers very low emissions with high thermal efficiencies and also can be readily adapted for CO_2 capture
- Currently, IGCC suffers from relatively high capital cost, low availability, and low fuel flexibility, in part because current generation of gasifier designs are based on past operating experience and 'cut-and-try' approaches rather than computational optimization of design and operation
- CFD design and analysis of gasifiers is limited by lack of knowledge of char gasification rates under practical T and pressure conditions
- Gasification kinetics are much more complicated than combustion kinetics, because of interaction of the two primary gasification steps and inhibition of these reactions by the reaction products

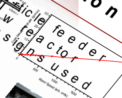
Approach: Char Gasification Kinetics

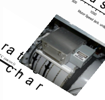
- Separate char formation (devolatilization) process from char gasification measurements to give 'cleaner' results on a common starting char material, especially since pressure influences volatiles yield and char structure
- Perform char gasification measurements with narrow char size cuts and low particle loading in gaseous environments
- Perform gasification measurements first in CO_2, then in steam in combination of CO_2 and steam, and then with some gasification products mixed in, to get definitive data for kinetics model development
- Model experimental results with detailed, porous particle model (SKIPPY) and with simple model to derive best kinetic expressions

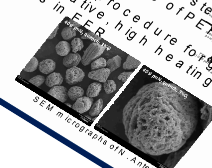
Results: Char Gasification Kinetics

- Developed and installed particle feeder for pressurized entrained flow reactor based on previous feeder designs used at MIT and Univ. of Utah
- Installed custom-designed steam generator on side of PEFR
- Developed procedure for generating representative, high heating rate char particles in FFR





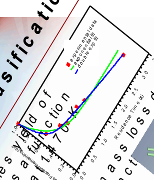




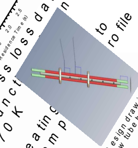
SEM micrographs of N. Antelope char

Results: Char Gasification Kinetics

- Measured volatiles yield of Pittsburgh coal as a function of residence time at 1470 K



- Collected gasification mass loss data for Pittsburgh char as function of residence time at 1470 K
- Redesigned PEFR heating to provide more uniform temp ro file at higher pressures



Design drawing of original flow tube heating system

Motivation: Pressurized Char Combustion Kinetics

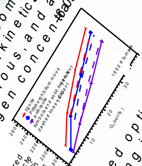
- Efficiency of both conventional and oxy-fuel combustion boilers is improved by operating at pressure: reduces gas cleanup losses and allows capture of water condensation enthalpy at higher temperature
- Furthermore, for CO_2 capture, pressurization of oxy-fuel boiler reduces additional compression requisite of CO_2
- Optimized design of pressurized burners and boilers requires quantitative kinetics for char combustion

Approach: Pressurized Char Combustion Kinetics

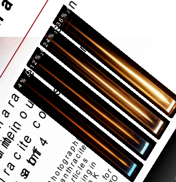
- Start with detailed study of char combustion kinetics at 1 atm for a wide range of coal rank for a wide range of [O_2] – this allows comparison to results at pressure with the same values of p_{O_2} (the provides differentiation of effects of partial pressure of oxygen and the total system pressure)
- Design and construct optical pyrometer for measuring individual char particle temperatures in the PEFR
- Collect optical data and burnout data for pc char for a wide range of pressures and p_{O_2}
- Model experimental results with detailed, porous particle model (SKIPPY) and with single particle model to derive best fit kinetic expressions

Results: Pressurized Char Combustion Kinetics

- Measured combustion chara[...] apparent kinetics of lignite, subbituminous, bituminous, and anthracite co[...] oxygen concentration [...] to 16 atm [...]

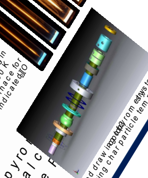


Measured char particle combustion temperatures



Photograph of anthracite particles burning in 1650 K furnace for indicated O_2

- Designed optical pyro[meter for] measuring individual c[har particle] temperatures in situ within the P[...]


Exploded drawing of pyrometry system for measuring char particle temperatures in PEFR

Archival Journal Publications

- A. Molina, J.J. Murphy, C.R. Shaddix, L.G. Blevins, "The Effect of Potassium Bromide and Sodium Carbonate on Coal Char Combustion Reactivity," *Proc. Comb. Inst.* 30:2187-2195 (2005)
- D.J. Maloney, E.R. Monazam, K.H. Casleton, C.R. Shaddix, "Evaluation of Char Combustion Models: Measurement and Analysis of Variability in Char Particle Size and Density," *Proc. Comb. Inst.* 30:2197-2204 (2005).
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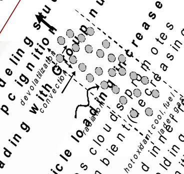
Extras

Ignition of Coal Particle Streams

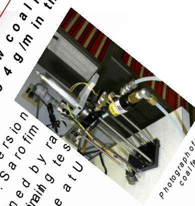
- Limited experimental and modeling studies of effect of particle loading on pc ignition
- Characterize particle loading with Group number

  $G \sim n_p r_p r_{cloud}$
- Competing effects as particle loading increases
  - presence of merged volatiles cloud 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

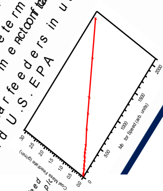


Ignition of Coal Particle Streams

- Enabled by installation of new coal particle feeder that prod... steady coal flow rates up to 4 g/min through small diameters tubing
  - design is modified version developed in Prof. Sarofim...
  - feed rate determined by ra... displacement ... ction of main... ng tes...
  - similar feeders in use at U... and U.S. EPA



Photograph of pulverized coal feeder



Coal feed calibration plo...