Pressurized Coal Pyrolysis and Gasification at High Initial Heating Rates

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Outline

- Review of the influence of pressure and heating rate on coal devolatilization
- Description of facilities
- Description of experimental findings
- Conclusions

Literature Review: Mass Release

- Effect of increasing pressure
 - Inhibits release of tar
 - More light gases produced by cross-linking reactions
 - Net decrease in volatiles
- Effect of increasing heating rate
 - Causes devolatilization to occur at higher temperatures
 - Higher rate of devolatilization
 - Higher yield of volatiles, especially tar
- CPD model predicts
 experimental trends



Shan G. PhD thesis, Department of Chemical Engineering, University of Newcastle (NSW), Australia, 2000.

Yu, J., J. A. Lucas and T. F. Wall, Progress in Energy and Combustion Science, 33(2), 135-170 (2007).

Literature Review: Swelling



Yu, J., D. Harris, J. Lucas, D. Roberts, H. Wu and T. Wall, *Energy & Fuels*, **18**(5), 1346-1353 (2004).

Yu, J., J. A. Lucas and T. F. Wall, Progress in Energy and Combustion Science, 33(2), 135-170 (2007).

Lee, C. W., R. G. Jenkins and H. H. Schobert, Energy & Fuels, 6(1), 40-47 (1992).

Literature Review: Swelling



- Effect of heating rate on swelling
 - Pressure effect mostly investigated at ≤10⁴ K/s

Gale, T. K., C. H. Bartholomew and T. H. Fletcher, *Combustion and Flame*, **100**(1-2), 94-100 (1995). Lee, C. W., R. G. Jenkins and H. H. Schobert, *Energy & Fuels*, **6**(1), 40-47 (1992).



Figure 3. Effect of pressure on maximum swelling volume (V_s) of Illinois No. 6 coal measured at 80 °C/min heating rate, in hydrogen and helium atmospheres.



Figure 4. Effect of pressure on maximum swelling volume (V_s) of Illinois No. 6 coal measured at 160 °C/min heating rate, in hydrogen and helium atmospheres.

Atmospheric Flat-Flame Burner (FFB)

- Advantages:
 - Char and soot formation at high heating rate ($\sim 10^5$ K/s)
 - Fueled by CH₄ or CO
 - Allows temperature flexibility (1100 K to 2000 K)
 - Adjust stoichiometry for % O_2 in post-flame zone
 - Very fast heat-up and shut-down times for ease of use
 - Residence time adjusted easily
- Disadvantages:
 - Limited to experiments at ambient pressure



Upgraded HPFFB

- Changed to up-flow
 - Reduces wear on the burner
- Probe moves to change residence time
 - Up to 800 ms for 1 section
 - Up to 1600 ms for 2 sections
 - Very short residence times available
- Operational pressures of 2.5-15 atm
 - Upgradeable to 30 atm
- Uses either CH₄ or CO with some H₂
 - Greater flexibility in gas composition
 - CO will not form soot
- Optical access available near burner
 - Check particle feeding
 - Limited optical particle velocities
- Faster startup
- Easier to disassemble



More Recent Upgrades

- Reduced burner diameter from 2" to 1"
 - Easier maintenance
 - Lower fuel costs
 - Closer match to collection probe ID
- New particle feeder
- More effective quench

15 atm HPFFB Temperature Profiles, Quench at 3"









Steam Gasification of Wyodak Coal

- Experimental Conditions
 - 2.5 atm
 - 1640 K, Φ=1.07
 - Fuel mixture
 - 84 mol% H₂ with CH₄
 - Gas composition
 - 27.4 mol % H₂O
 - 3.0 mol % CO₂
 - 1.8 mol % H₂
 - 0.06 mol % O₂
 - $-77 \,\mu m$ particles



Steam Gasification of Wyodak Coal

90 ms char fully pyrolyzed CPD predicts 62% MR_{daf} in ~30 ms Little change from 210-870 ms Linear gas temperature decrease of ~300 K from peak over 14 inches (660 ms) Highly porous chars - Coal $\rho_{\rm p} = 1.49$ N_2 surface area of 360 m²/g at 210 ms Zone II behavior near burner - d_p and ρ_p changing in first 210 ms Zone III calculations predict 100% conversion ~30 ms after full pyrolysis





CO₂ Gasification of Wyodak Coal

- **Experimental Conditions**
 - 2.5 atm, 5 atm, 10 atm, 15 atm
 - Probe set 0.75" above burner
 - 1700 K, Φ=1.24-1.27
 - 5 atm, 15 atm
 - Probe set 3" above burner
 - 1700 K, Φ=1.24-1.27
 - 1900 K, Φ=1.15
 - Fuel mixture
 - 92 mol% CO with H₂
 - Gas composition at 2.5 atm
 - 1.8 mol % H₂O
 - 15.7 mol % CO₂
 - 11.9 mol % CO
 - 0.4 mol % H₂





44 ms

CO₂ Gasification of Wyodak Coal



Pressure (atm)

Condition	2.5 atm, 40 ms, 1700 K	5 atm, 40 ms, 1700 K	5 atm, 120 ms, 1700 K	5 atm, 120 ms, 1900 K	10 atm, 40 ms, 1700 K	15 atm, 40 ms, 1700 K	15 atm, 120 ms, 1700 K	15 atm, 120 ms, 1900 K
ρ _p (g/cc)	0.83	0.89	0.91	0.67	0.98	0.93	0.91	0.79
d/d _o	0.91	0.90	0.86	0.96	0.89	0.91	0.89	0.93
Ash Tracer MR (daf %)	46.8	46.6	55.1	60.2	44.5	46.1	50.4	53.9
Average Al/Si Tracer MR (daf %)	52.3	51.3	57.4	57.0	47.4	46.9	52.4	52.4

CO₂ and H₂O Gasification at 2.5 atm, 1700 K



Wyodak Elemental Changes



Atmospheric Swelling

- U.S. bituminous coal
- Atmospheric FFB
 - Varied particle size to change heating rate
- Swelling trends consistent with previous work
 - Sharp decrease between 10⁴ - 10⁵ K/s
 - Apparent asymptote of ~0.9 above 10⁵ K/s
 - Eiteneer data indicate maximum swelling occurs slightly below 10⁴ K/s



Eiteneer, B., et al., 26th Annual International Pittsburgh Coal Conference, Pittsburgh, PA (2009).

Gale, T. K., C. H. Bartholomew and T. H. Fletcher, Combustion and Flame, 100(1-2), 94-100 (1995).

Zygourakis, K. Energy & Fuels 7, 33-40 (1993).

U.S. Bituminous Coal A Swelling

Particle Size (µm)	149-177	88-105	53-66	
Heating Rate (K/s)	$4.1 \cdot 10^4$	$7.2 \cdot 10^4$	$2.0.10^{5}$	
MR (% daf)	60.29	63.25	61.44	
ρ/ρ ₀	0.24	0.39	0.53	
d/d ₀	1.22	1.00	0.93	







10 atm U.S. Bituminous Coal B



1 gram coal fed

Cenospheric char particles fragile
Char accumulates in horizontal cyclone –Must empty cyclone frequently to avoid fragmentation

0.5 gram coal fed



Planned Gasification Studies

- Extend HPFFB subbituminous data set
 Longer RT, multiple temperatures
 - Collect HPFFB bituminous coal data
- Determine mass release
 - Compare to CPD code, make adjustments
 - Optimize gasification kinetic parameters
- Measure structural information
 - Determine and model swelling (correlation for CPD)
 - N₂ and CO₂ surface areas
 - Extent of fragmentation
- PTGA gasification kinetics of HPFFB chars
 - Compare kinetics derived from PTGA and HPFFB
 - Make adjustments to gasification model

Summary and Conclusions

- Modified HPFFB suitable for gasification studies
 - Heating rates of $\sim 10^5$ K/s at up to 15 atm
 - Gas composition, residence time more flexible
- Preliminary gasification experiments
 - Subbituminous coal in Zone II conditions
 - High surface area and porosity in steam gasification
 - Pyrolysis yield decreases with increasing pressure
- Atmospheric swelling experiments
 - Confirms previous trends
 - Reinforce suggestion of swelling ratio < 1 at heating rates of ~10⁶ K/s

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15 atm Bituminous Coal B with Soot Agglomerates



BYU HPFFB



Wyodak Ultimate Analysis (daf)

Sample	С	H	Ν	S	O (diff)
Wyodak Coal	72.25	5.30	0.94	0.50	21.01
2.5 atm 90 ms char (H ₂ O, 1640 K)	91.14	1.11	1.06	0.27	6.42
2.5 atm 210 ms char (H ₂ O, 1640 K)	92.50	1.16	0.86	0.39	5.10
2.5 atm 870 ms char (H ₂ O, 1640 K)	92.69	1.22	0.94	0.53	4.62
2.5 atm 33 ms char (CO ₂ , 1688 K)	86.99	2.08	1.31	0.44	9.18
5 atm 38 ms char (CO ₂ , 1714 K)	89.38	1.78	1.33	0.41	7.10
10 atm 41 ms char (CO ₂ , 1724 K)	90.07	1.73	1.29	0.43	6.48
15 atm 44 ms char (CO ₂ , 1695 K)	91.38	1.50	1.35	0.41	5.37
5 atm 124 ms char (CO ₂ , 1714 K)	91.54	1.19	1.40	0.58	5.30
5 atm 110 ms char (CO ₂ , 1881 K)	91.84	0.98	1.35	0.72	5.10
15 atm 115 ms char (CO ₂ , 1695 K)	91.26	1.16	1.33	0.50	5.74
15 atm 120 ms char (CO ₂ , 1953 K)	90.95	1.14	1.26	0.64	6.01