GASIFICATION KINETICS OF INDIANA COALS IN THE LOCATIONS PROMISING FOR UCG TECHNOLOGIES

Final Report
to the
Indiana Center for Coal Technology Research (CCTR)

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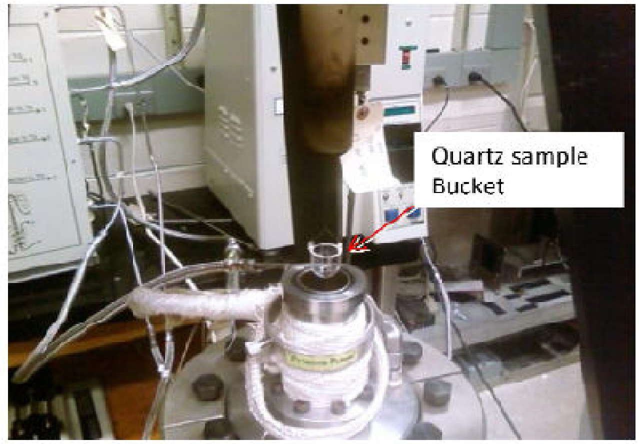
1. Motivation and Objectives

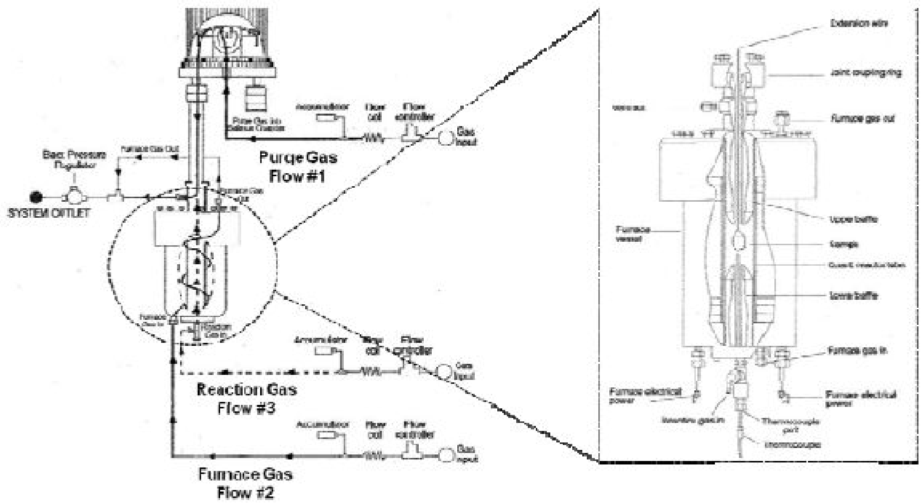
Our prior CCTR-funded project on underground coal gasification (UCG) has shown that this technology has great potential to grow and replace/complement traditional methods for coal mining and surface gasification [1]. Some basic screening criteria for selecting UCG sites in Indiana were formulated in the previous report [2]. Using the available information on the characteristics of coal seams in Indiana, it was recommended to focus on the Springfield and Seelyville coal beds. For these coals, maps were generated that show thickness, depth and other characteristics, such as moisture content and heating value for these two coals. Analysis of the maps and additional information identified nine promising zones for UCG in Indiana (five for the Seelyville and four for the Springfield Coal). For these zones, the lists of characteristics, including the amounts of available coal, were prepared. Based on these data, preliminary recommendations on the future selection of a suitable location for UCG operations were made. As a follow-up of the first project described above, we have completed the second CCTR-funded project which investigates gasification kinetics of coal samples from the Seelyville and Springfield seams. This investigation is required because Indiana coals, although of similar rank, may have significantly different chemical and petrographic compositions, also accompanied by differences in porosity and permeability of the coal [3]. Knowledge of gasification kinetics and understanding of the permeability effect is necessary for the modeling and success of UCG operations. In the next section, the results obtained in the current project are described.

2. Accomplishments of Current Project

2.1. Experimental Setup

In general, the UCG process is operated at elevated pressure which is determined by the depth of coal seam. For this reason, the gasification kinetics were investigated using a Pressurized Thermogravimetric Analysis (P-TGA) facility (Cahn Instruments, Model TG-151), available in Purdue's School of Chemical Engineering. Figure 1 shows a photograph and schematic of the P-TGA setup. It consists of three gas chambers: the reactor chamber, balance chamber and the furnace chamber. The furnace chamber is separated from the reactor chamber by a quartz tube. The controller with its built-in mass flow control (MFC) maintains the flow rates of the three gases (i.e. furnace, reaction, and purge gas). Either about 100 mg single coal particle (5 mm cubic-shape) or fine coal particle (63-75 μm) sample was placed on the sample bucket inside the reactor. A back-pressure regulator is used to control the reactor pressure. Starting at room temperature, with programmed heating rate (10 ^oC/min), the reactor was maintained for 10 minute hold at 150 ^oC to remove any moisture in the sample. Following this, it was heated to 1000 ^oC at the same heating rate. The profiles of temperature and sample weight were recorded at 1-second interval. Note that buoyancy effect and drag force to sample weight were observed at high pressure conditions, which have been reported in the prior literature. Thus, some blank tests were also carried out to compensate for these effects from the kinetic results.





2.2. Properties of Coal

The chemical composition of coal is defined in terms of its proximate and ultimate analyses. The parameters of proximate analysis are moisture, volatile matter, ash, and fixed carbon, while ultimate analysis encompasses the quantitative determination of carbon, hydrogen, nitrogen, sulfur, and oxygen. The heating value of coal is the heat liberated by its complete combustion with oxygen. Table 1 shows proximate and ultimate analyses of the two coal samples, along with their heating values and depth of each coal seam [4]. Further, coal is classified into four general ranks: lignite, sub-bituminous, bituminous and anthracite, reflecting the progressive response of individual deposits of coal to increasing heat and pressure. Bituminous type is the most plentiful form of coal in the U. S. and, in general, has carbon content ranging from 45 to 86 percent and heating value 10,500 to 15,500 BTU/lb. As shown in Table 1, both coal samples used in this study show the typical properties of bituminous coal.

Table 1. Properties of coal samples

Coal type	Proximate Analysis (dry basis)			Ultimate Analysis (dry basis)						Heating Value (Btu/lb)	Depth (ft)
	Moisture* (%)	Volatile Matter (%)	Fixed Carbon (%)	Ash (%)	C (%)	H (%)	N (%)	O (%)	S (%)		
Seely-ville	8.0	40.6	47.8	11.6	69.7	4.9	1.5	7.6	4.8	12821	640
Spring-field	6.2	39.1	55.8	5.2	78.6	5.2	1.7	8.4	0.9	13763	899

*as received

2.3. Results of Pyrolysis/Gasifcation Kinetics

As noted above, two coal samples from Seelyville and Springfield seams were tested in this study. The effects of various conditions (i.e. temperature, pressure, O_2 concentration, steam concentration, coal particle size, etc.) were investigated.

Before describing the experimental results, we briefly note the phenomena that occur when a coal sample is heated in an inert atmosphere (N_2 in this study). First, moisture is removed starting at 100 ^{o}C (Zone 1), which is followed by release of volatiles that evolve at a higher rate between 400-600 ^{o}C (Zone 2), and then at a slower rate. If steam is present, then for temperatures ≥ 600 ^{o}C, char gasification occurs accelerating weight loss (Zone 3). The typical experimental results of coal pyrolysis/gasification in the absence and presence of steam are shown in Figure 2.

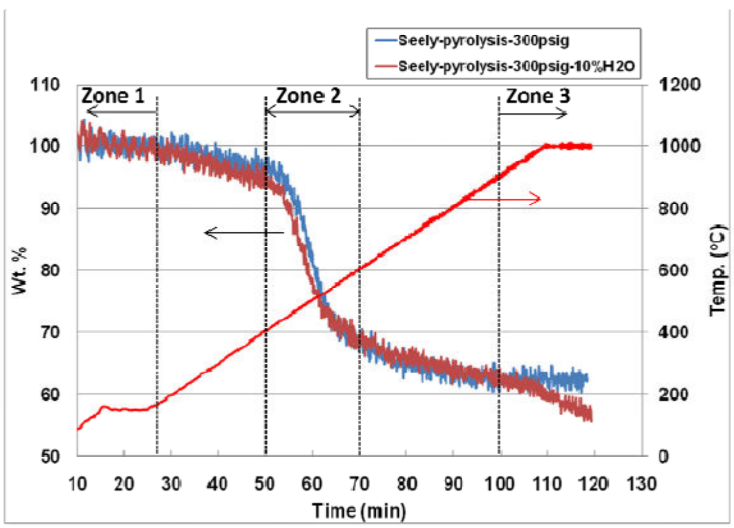


Figure 2. Typical behavior of coal pyrolysis and gasification with steam (Seelyville coal)

Table 2 shows the calculated kinetic parameters from the TGA thermograms under pyrolysis conditions (T < 600 ^{o}C). The kinetic parameters, activation energy (*E*) and pre-exponential factor (A), were calculated assuming first-order reaction following the Coats-Redfern method, which has been widely used for kinetic studies with TGA [5].

Table 2. Kinetic parameters of coal pyrolysis/gasification at different conditions

Coal type	Test conditions	Temp. (°C)*	*E* (kJ mol^{-1})	A(s^{-1})	R^2 value
Seelyville	1 atm	476-570	67.5	1.26E+03	0.97
	100 psig	467-573	72.6	2.35E+03	0.97
	[a)]300 psig	476-560	83.3	2.14E+04	0.93
	100 psig -10%H_2O	465-574	71.1	1.64E+03	0.97
	[a)]300 psig-10%H_2O	431-542	72.7	3.33E+03	0.96
	[a)]300 psig-[c)]Fine-10%H_2O	421-519	58.7	2.62E+02	0.94
Springfield	1 atm	478-580	61.8	3.90 E+02	0.95
	100 psig	470-563	66.2	6.82E+02	0.98
	[b)]400 psig	430-521	85.8	4.12E+04	0.94
	100 psig -10%H_2O	459-576	51.8	5.87E+01	0.98
	[b)]400 psig-10%H_2O	434-529	70.2	2.73E+03	0.94
	[b)]400 psi-[c)]Fine-10%H_2O	446-522	61.9	4.97E+02	0.91

[a), b)] corresponding to pressure at each coal seam sample, [c)] fine particle coal (63-75 µm)

The parameters were calculated for different pressures (1 atm to 400 psig) under pyrolysis conditions (with and without steam). In particular, 300 and 400 psig correspond to the pressures at the depths of Seelyville and Springfield coal seams, respectively.

Figure 3 shows the effect of pressure on pyrolysis reaction in the absence of steam. It was found that the activation energy and pre-exponential factor increase with increasing pressure, as shown in Table 1. The reason for this is likely that release of volatile materials from coal is suppressed at higher pressures.

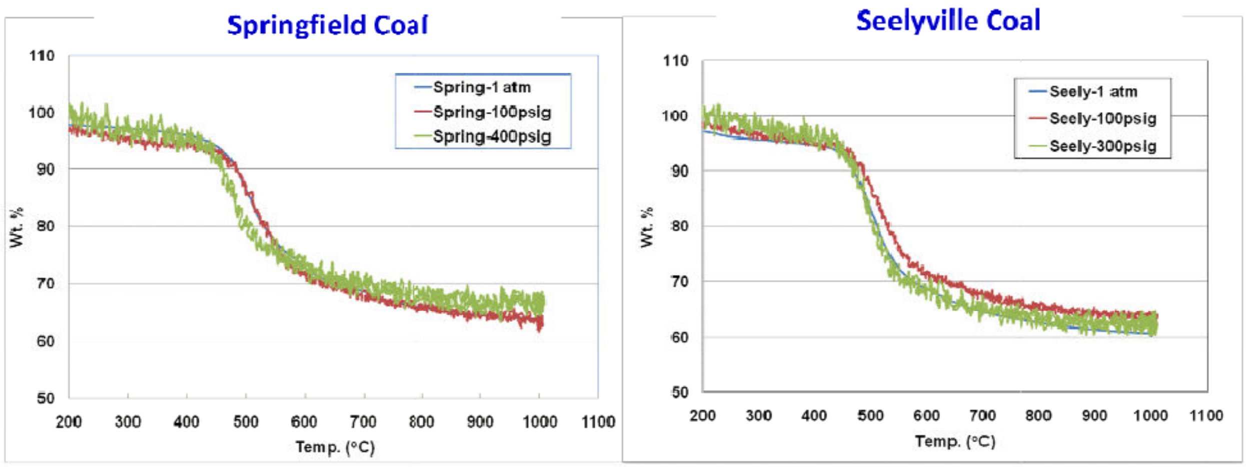


Figure 3. Effect of pressure on coal pyrolysis

Table 2 and Figure 4 also show the effect of steam on pyrolysis/gasification kinetics. Steam (10 volume % in flowing N_2) was introduced into the reactor by syringe pump through heated tubing at 100, 300, and 400 psig. It was found that the activation energy decreases, while the amount of thermal decomposition increases with addition of steam. The most significant effect was observed for temperature above 900 °C. It is clear that the presence of water vapor enhances coal decomposition by introduction of the gasification process ($C + H_2O \rightarrow CO + H_2$).

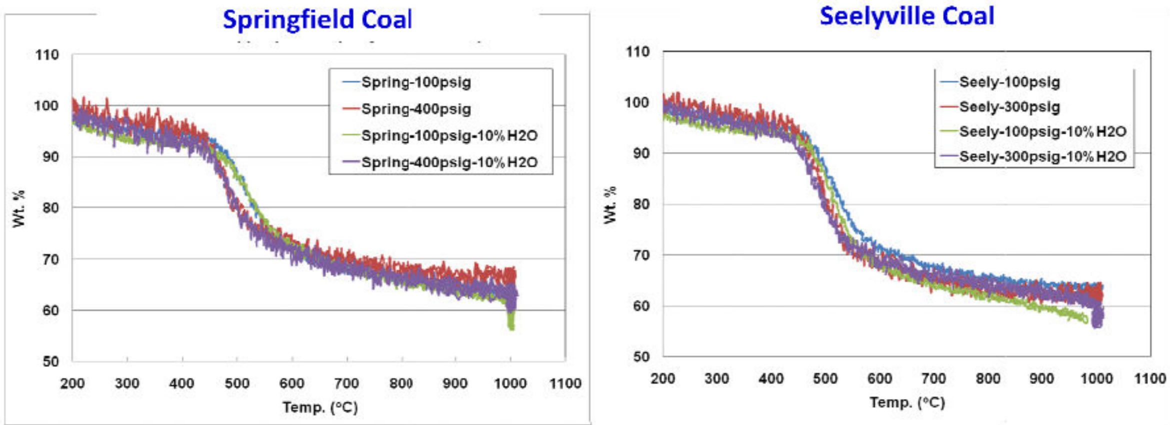


Figure 4. Effect of steam on coal pyrolysis and gasification

The effect of particle size on pyrolysis/gasification reaction was also investigated in this study. It was found that fine particle coal shows higher reaction rates than the larger single coal particle sample and also has lower activation energy, as shown in Figure 5.

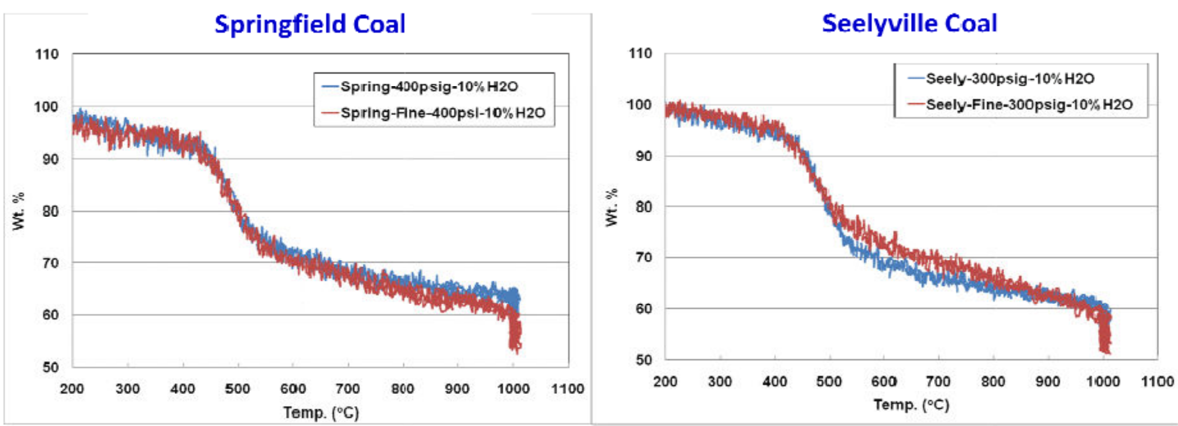


Figure 5. Effect of coal particle size on pyrolysis/gasification reaction

When oxygen is present, coal decomposition is accelerated by its combustion. The effect of pressure under combustion atmosphere (21% O_2 concentration) is shown in Figure 6. With increasing pressure, the temperature for the maximum reaction rate is shifted to lower values and weight change rate increases. This occurs because mass transfer of oxygen into the coal particle increases with pressure, which enhances the combustion rate.

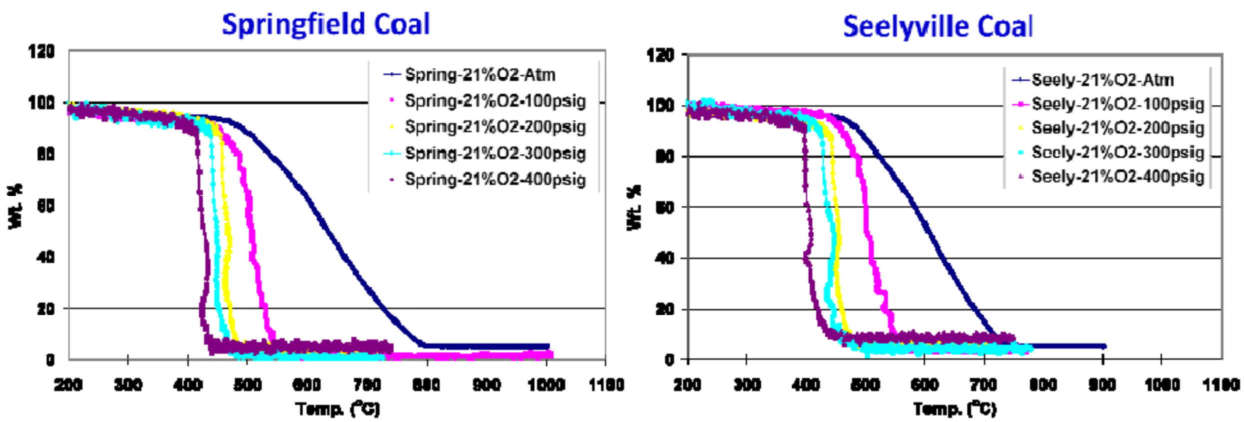


Figure 6. Effect of pressure on coal pyrolysis/combustion reaction

When oxygen and steam are present, then both combustion and gasification can occur. Figure 7 shows this effect for the Seelyville coal. It is clear that the presence of water in oxygen enhances coal decomposition.

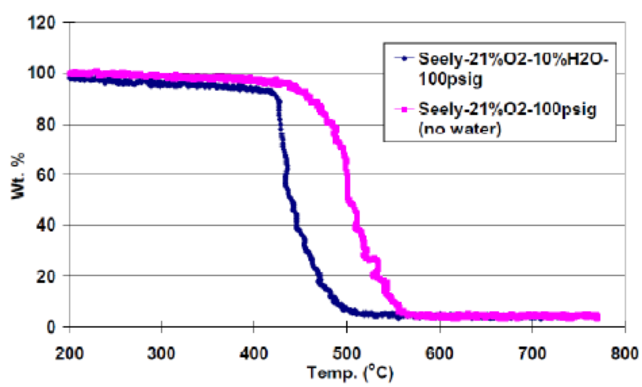


Figure 7. Effect of steam on combustion/gasification reaction for Seelyville coal

3. Summary

In summary, the effects of operating conditions on pyrolysis/gasification process for two coals from Seelyvlle and Springfield seams were investigated. Both coals exhibited similar trends in thermogravimetric analysis. The Springfield coal, however, had lower activation energy as compared to the Seelyville coal. In future work, analysis of the produced gas composition will be required to identify the gasification characteristics of the coals. Based on the results obtained in this project, a proposal titled "Laboratory-Scale Underground Coal Gasification Studies Using Selected Indiana Coal Samples" has been submitted for funding to CCTR [6].

4. References

[1] Shafirovich, E., and Varma, A., "Underground Coal Gasification; A Brief Review of Current Status," *Industrial and Engineering Chemistry Research*, **48**, 7865-7875, 2009.

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[4] Mastalerz, M., Personal communication, December 2009.

[5] Coats, A. W. and Redfern, J. P., "Kinetic Parameters from Thermogravimetic Data," *Nature*, **201**, 68-69, 1964.

[6] Varma, A., "Laboratory-Scale Underground Coal Gasification Studies Using Selected Indiana Coal Samples," Proposal to the Indiana Center for Coal Technology Research (CCTR), November 29, 2010.